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RADIOLOGICAL SIGNIFICANCE OF THORIUM PROCESSING IN MANUFACTURING

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ABSTRACT

The study of thorium processing in manufacturing comprised monitoring programs at a plant where thorium dioxide was in use and another where the use of thorium nitrate had been discontinued. The measurements of the solubility in simulated lung fluid proved that both materials belonged in the Y Class with dissolution half-times greater than 500 days. Bioassay measurements of 20 subjects from both facilities proved that in vitro monitoring methods, urine, feces, hair and nails analysis were not sufficient indicators of thorium uptake. In vivo monitoring by phoswich and large sodium iodide detectors were proven to be good methods of determining thorium lung burdens. The thoron in breath technique was shown to have a lower limit of sensitivity than lung counting, however, due to lack of information regarding the thoron escape rate from the thorium particles in the lungs the method is not as accurate as lung counting. Two subjects at the thorium dioxide facility had lung burdens of 21± 16 Bq and 29± 24 Bq Th^{232} and one at the thorium nitrate facility had a lung burden of 37± 13 Bq. Improvements in the procedures and use of a glove box were among the recommendations to reduce the inhalation of thorium by workers at the thorium dioxide facility. Decontamination of several rooms at the thorium nitrate facility and sealing of the walls and floors were recommended in order to reduce the escape of thoron gas into the room air. The risk to non Atomic Radiation Workers was primarily due to thoron daughters in air while gamma radiation and thorium in air were less important. Conversely, at the thorium dioxide facility the inhalation of thorium in air was the most significant exposure pathway.

RÉSUMÉ

L'étude sur l'utilisation du thorium dans les usines de fabrication comprenait des programmes de surveillance dans une usine où l'on utilisait du bioxyde de thorium et dans une autre où l'on avait cessé d'utiliser du nitrate de thorium. Les mesures de solubilité dans du fluide pulmonaire simulé ont prouvé que les deux matières appartenaient à la classe Y et qu'elles mettaient plus de 500 jours pour être à moitié dissoutes. Les relevés pris au cours d'essais biologiques menés auprès de 20 sujets provenant des deux installations ont prouvé que les méthodes de surveillance in vitro et que l'analyse d'urine, des fèces, des cheveux et des ongles ne pouvaient servir d'indicateurs suffisants d'incorporation de thorium. La surveillance in vivo par des scintillateurs «sandwich» (phoswich) et de grands détecteurs à l'iodure de sodium s'est avérée une bonne méthode pour déterminer la charge de thorium dans les poumons. La technique du thoron dans l'haleine a montré qu'elle possédait un seuil de sensibilité inférieur au comptage pulmonaire, mais, à cause du manque de renseignements au sujet du taux de fuite du thoron qui s'échappe des particules de thorium dans les poumons, la méthode n'est pas aussi précise que le comptage pulmonaire. Deux des sujets provenant de l'installation de bioxyde de thorium avaient des charges pulmonaires de 21± 16 Bq et de 29± 24 Bq Th^{232} et un des sujets provenant de l'installation de nitrate de thorium avait une charge pulmonaire de 37± 13 Bq. On a recommandé, entre autres, d'améliorer certaines procédures et d'utiliser une boîte à gants pour réduire l'inhalation de thorium par les travailleurs de l'installation de bioxyde de thorium. On a recommandé de décontaminer plusieurs salles et de sceller les murs et les planchers à l'installation de nitrate de thorium afin de réduire la fuite de thoron dans

l'atmosphère des salles. Le risque pour les personnes qui ne sont pas des travailleurs sous rayonnements était dû avant tout à la présence de produits de filiation du thoron dans l'air, mais le niveau de rayonnements gamma et de thorium dans l'air était moins important. Inversement, c'est l'inhalation de thorium dans l'air qui représentait la voie d'exposition la plus importante à l'installation de bioxyde de thorium.

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1.0 INTRODUCTION

This report summarizes the work carried out during the study for the Atomic Energy Control Board (AECB) titled 'The Radiological Significance of Thorium Processing in Manufacturing'.

1.1 Background

Thorium compounds, mainly in the form of powders, are used in the manufacture of numerous commercial products such as optical glass, gas mantles, tungsten welding electrodes, electrical filaments and in various metal alloys. Many of these compounds can be inhaled resulting in radiological insult to workers and, when released to the environment, to the public.

1.2 Objective

The objective of the study is to assess the radiological significance of radionuclides associated with thorium processing in the manufacture of industrially and commercially used devices in Canada.

1.3 Scope Of Work

The scope of work includes an identification of the dosimetrically significant physical and chemical properties of thorium compounds handled and produced in Canadian industry. An assessment of the radiological significance of radionuclides associated with thorium processing will follow by means of:

a) a review of available information;
b) monitoring in the workplace of the levels of radionuclides associated with thorium processing in manufacturing;
c) a program of in vitro and in vivo measurements (urinalysis, body burden, thoron in breath) of workers handling these compounds; and
d) a study of the solubility in simulated lung fluid of the thorium compounds handled and stack emissions from the manufacturing plant.

1.4 Tasks To Be Performed

The tasks to be performed include:

a) identify the thorium compounds processed, the quantities handled, and thorium by-products in stack emissions in the manufacture of industrially and commercially used devices in Canada;

b) describe the physical and chemical properties of the radioactive compounds associated with the thorium processing;
c) assess the radiological significance of these compounds by:
   i) a review of available information
   ii) monitoring of the levels of the radioactive compounds in the workplace and outside (where there are stack emissions)
   iii) carrying out a program of in vitro and in vivo measurements for workers if warranted by ii) above (as determined by the Scientific Authority of the AECB)
   iv) carrying out a study of the solubility in simulated lung fluid of the thorium compounds handled and in stack emissions; and
   v) relating the information obtained in i), ii) and iii) to the radiological significance for workers and the public by the use of existing metabolic and dosimetric models for radio-compound uptakes, and environmental pathway analysis.

1.5 Summary

The study of thorium processing in manufacturing comprised monitoring programs at one plant where thorium dioxide was used and at another where there were residual effects after the use of thorium nitrate had been discontinued.

Solubility in simulated lung fluid measurements were made using materials from both facilities. The results placed both the thorium dioxide and the thorium nitrate in the Y Class with solubility half-times greater than 500 days. Although the thorium nitrate was highly soluble in the nitrate form, immediately after going into solution the thorium formed an insoluble complex in the simulated lung fluid.

Bioassay measurements of a total of 20 subjects from the two facilities illustrated that in vitro monitoring methods, urine, fecal, hair and nails analysis were not sufficiently sensitive indicators of thorium uptake. Lung burdens were measured in three subjects at 21$\pm$ 16 Bq, 29$\pm$ 24 Bq and 37$\pm$ 13 Bq by in vivo methods using phoswich and large sodium iodide detectors. Lung burdens were detected with a lower limit of sensitivity by the thoron in breath technique than by lung counting. However, due to uncertainty about the thoron escape rate from thorium particles in the lungs, the thoron in breath technique cannot be used to accurately calculate lung burdens with the limited amount of information available.

Extensive monitoring of the occupational and surrounding environment was carried out at the two plants. Gamma and beta exposure rates were measured, thoron, thoron daughters and Th^{232} were analysed in air using open face and respirable air samplers. Th^{232} was measured in soil, concrete and roofing material, total and removable alpha contamination was measured and air velocities and flow rates were measured. Some aspects of the radiological significance of the findings were distinctly different at the two plants. At Facility A the risk to non Atomic Radiation Workers was primarily due to thoron daughters in air while gamma radiation and Th^{232} in air were less important. On the other hand, at Facility B the inhalation of Th^{232} is

the significant exposure pathway. Recommendations regarding the immediate remedial action to be taken at Facility A centered on decontaminating three of the most highly contaminated areas and sealing the walls and floors to decrease the escape of thoron gas into the room air. In the longer term when renovations are made or the property is sold, it will be necessary to breakup and dispose of the thorium impregnated concrete, soil and roofing material in approved sites. Recommendations regarding improvements in procedures and equipment were made to reduce the inhalation of thorium by the workers at Facility B. Until it has been demonstrated that thorium uptakes have been reduced it is recommended that semi-annual lung counting and thoron in breath measurements be made on each subject working in the mixing and coating rooms.

Contact beta-gamma exposure rate measurements were made using thermoluminescent dosimeters (TLDs) held against two types of lantern mantles, pieces of thorium metal and electrodes onto which thorium had been mechanically fixed. Appropriate filtration was used so that the results were applicable to finger and hand exposures with and without cotton gloves. The contact exposure rates ranged from 3.5 to 4.8 mR/h for the mantles, 17.3 to 30 mR/h for the thorium metal and 0.23 to 0.63 mR/h for the electrodes.

2.0 IDENTIFICATION OF QUANTITIES AND PROPERTIES

At both facilities under study relevant information concerning all aspects of the use of thorium was collected.

2.1 Background Information From Facility A

Thorium was used in manufacturing at Facility A between 1947 and 1983. The stock material is thorium nitrate $\{Th(NO_3)_4.4H_2O\}$ which is a colourless crystalline material with molecular weight of 552.12. It is very soluble in hot or cold water and alcohol and is slightly soluble in acetone. Thorium has been purchased from May & Baker Canada Inc. since the late 1970's. Fifty kilogram lots of the stock material were packaged in double polyethylene bags with sealed tops and shipped in steel drums. The drum lid was secured with a hoop.

The thorium usage rate during recent years is reported to have been:

Year	kg Per Year
1977	33,700
1980	11,930
1981	16,930
1982	9,780
1983	2,370

As manufacturing demanded, thorium powder was removed from the storage room to the wet process area where it was dissolved in

water. The mantle materials were soaked for about an hour in the thorium solution and then centrifuged to remove the excess. The materials were then spread on racks and air dried for about an hour in ovens. Then the materials were transferred to a cabinet and exposed to anhydrous ammonia to convert the thorium nitrate to the oxide form. The materials were washed and rinsed to remove excess ammonia and then they were centrifuged and hung on racks to dry. In order to stiffen the materials a lacquer was applied and dried.

In the dry processing area, the thorium laden materials were sewn, clipped, stamped with the manufacturers name and a hardening solution was applied to one end of the product. Until 1979 the hardening solution contained thorium, but it has not been an ingredient of the hardening solution since that time. Finally, the mantles were stacked on wooden trays to dry.

Stock thorium nitrate was stored in a small metal shed located external to the main manufacturing building on the ground floor. Controlled access was through an exterior door. Ventilation was not provided for this area. The inventory fluctuated with use and delivery of stock with a normal maximum of 30 drums. Until about 1970 the wet and dry process areas were on the main floor of the manufacturing building. After 1970 the two processes were moved to the two second floor rooms that were in use until thorium processing was discontinued in 1983. Solid thorium bearing waste was disposed of with normal garbage or burned on the premises until 1981. Solid thorium waste generated since that time was held in steel drums and then disposed of at the Radwaste Disposal site at Chalk River.

Thorium bearing liquid waste resulted from normal cleaning of the manufacturing areas and the process itself. Machines in the dry process area were steam cleaned once per week and the floor in the wet process area was washed each night. All waste water was drained directly to the city sewers. Process solutions also passed directly to the city sewers from the centrifuge, washer, sink and four floor drains in the wet process area.

Exhaust air from the drying ovens, denitriding and lacquering cabinets is vented directly to the roof. Exhaust air from the processing area is also vented directly to roof exhaust stacks.

AECB and company files show that external dosimetry and urinalysis records were maintained for workers in the early 1970's. Between 1973 and 1979 both monitoring methods appear to have been discontinued. In 1979 external dosimetry and urinalysis monitoring were reinstated and recently (since 1980) thorax burden measurements have been made by the Radiation Protection Bureau in Ottawa. For those subjects who participated in this study, work histories are given in Table 2-1.

2.2 Background Information From Facility B

Thorium in both metallic and oxide forms has been in use at Facility B since at least 1966. The thorium dioxide {ThO_2, molecular weight 264.02} comprises fine white crystals and is insoluble in water, dilute acid and alkaline solutions. It is soluble in hot sulphuric acid. Thorium dioxide stock material is currently supplied by May & Baker Canada Inc. in 10 kg lots sealed in a polyethylene bag in a small drum. The drum lids are secured with a hoop.

The thorium metal is procured in sheets 0.076 mm thick and is cut on site into pieces 2.5 x 0.25 mm and weighing 0.5 mg. Thorium metal has a molecular weight of 232.038 and a density of 11.7 g/cm^3. It is insoluble in water and only slightly soluble in nitric acid. It is soluble in hydrochloric and sulphuric acids.

Eleven subjects, who have worked with thorium at Facility B during some time since 1966, volunteered to participate in the study. Their job descriptions and work histories are summarized in Table 2-2.

Complete and reliable records of thorium dioxide preparation have been kept by subject B7 since 1966. The annual rate of thorium usage has been taken from those records and is reproduced in Table 2-3.

Current handling of thorium takes place in four locations; the mixing room, the coating room, the manufacturing area and the storage room. Thorium dioxide stock material is stored in a glove box in the mixing room and as production demands it is prepared in solution currently about once every two months. The solution is a mechanical mixture of thorium dioxide and bicarbonates in methanol. The mixture is crushed in a ball mill and then transferred into polyethylene bottles for storage. The solution is stored in the mixing room until required and the bottles are rolled continously to maintain the mix.

In the coating room the thorium is mechanically fixed onto electrodes on a daily basis. Thorium/methanol mixture from a stock of a few litres kept in the coating room is poured into small trays and the electrodes are immersed in the liquid. The trays are transferred into a vacuum chamber and the air pressure is reduced to about 5 cm of mercury to release trapped air. After the air pressure has been raised to normal the trays are transferred to a water bath where the methanol is evaporated leaving dry thorium dioxide and coated electrodes. The electrodes are transferred to a clean tray after the excess thorium is shaken off. Instructions require that coating room operators wear rubber gloves, use the fume hood for most tasks and no smoking or eating is allowed.

In the manufacturing area the electrodes are fired in a hydrogen furnace to fix the thorium before use. The final assembly stage involves two steps: a) the electrodes covered with thorium are handled individually with tweezers and are welded to a second component, b) the welded components are sealed inside the final assembly by a second worker.

The present coating room has been in use since 1978. From about 1965 until 1978, the coating room was located on a lower floor.

All solid thorium waste materials are presently collected and stored separately in specially designated barrels. These barrels are held in the area set aside for storing hazardous solvents and reagents.

The thorium metal sheets received from the supplier are about 4 cm on a side and are 0.076 mm thick. Once per month the sheets are cut into narrow strips and cleaned by electrolysis. After cleaning they are cut into 0.5 mg pieces and are stored in a nitrogen atmosphere until required. Instructions require that the thorium be handled by tweezers or with cotton gloves.

The air drawn from the mixing room and the fume hoods therein have a common exhaust on the north wall of the building. Air is drawn into the room around the door by the negative pressure generated by the exhaust fans.

In the coating room there is a supply of fresh air provided which greatly exceeds the exhaust drawn from the room through the hooded box in which the dry thorium powder is handled. Therefore, some of the coating room air is expelled through the doorway into the manufacturing area by the positive pressure created inside the room. An exhaust fan draws air through a filter from the hood in the coating room and exhausts it through a roof vent which is part of an adjacent ventilation system.

3.0 REVIEW OF RADIOLOGICAL DATA

A literature survey was conducted using the International Nuclear Information System (INIS) which was accessed via the National Research Councils CAN/OLE database system. INIS is maintained and updated by the International Atomic Energy Agency and covers all peaceful applications of atomic energy. The database contains reports covering the period 1970 to the present.

Abstracts and publication data of the reports dealing with occupational, environmental, monitoring and dosimetric aspects of thorium and its daughters are included in a bibliography which is available in the AECB's library in Ottawa, Ont.

No references addressing the solubility of thorium or its compounds in simulated lung fluid were found. Several additional reports relating to the studies of thorotrast patients were identified.

Rundo (Ru78) has published a review of the radioactive properties and biological behaviour of thorium^{232} and its daughters. The thorium decay chain, Figure 3-1, comprises isotopes of eight different chemical elements, three are represented twice. As a result of nuclear transformations, the chemical identity of the radioisotopes changes. When these radioisotopes are located inside living organisms, the new chemical species may be incompatible with the site environment.

The uptake of thorium dioxide by inhalation and injection into beagle dogs was studied by Ballou and Hursh (Ba72). The dust was enriched in Th^{228} to enhance thoron production and measurement sensitivity. Airborne dust concentrations averaged 188 mg/m^3 with a particle size distribution of 0.11 µm CMD (count median diameter-median diameter based on number of particles) σg 2.13 and 1.75 µm AMMD (aerodynamic mass median diameter) σg 2.86. Immediately after inhalation the dust burdens averaged 2.12 mg ThO_2; but only an average of 0.27 mg was located in the lower respiratory tract. The thoron/Th^{228} ratio in lung varied from 0.10 to 0.17 during the interval from 3 to 150 days following inhalation; indicating a steady-state relationship was quickly established. Higher values have been reported for human thorotrast patients, however, the ThO_2 particles were much smaller at 75 Å CMD for thorotrast compared to 2000 Å in Ballou's work. Ballou also measured a biological half time in the lungs of 350 - 500 days which agrees well with the ICRP Task Group on Lung Dynamics recommendations.

Recently Newton (Ne81) reported a case where a male became internally contaminated with Th^{228}, presumably through inhalation of the oxide. A seven year study of this subject resulted in the conclusion that the biological half-life for long term clearance from the body was at least 14 years and probably longer. Also, it was concluded that the material had cleared the lung as a Class W compound. However, this was not observed directly since the detection of the burden and subsequent monitoring took place after the lungs had been cleared.

4.0 FACILITY MONITORING

4.1 Methods And Calibrations

4.1.1 Gamma Exposure Rate Surveys

Gamma exposure rate measurements were made using a 2" x 2" sodium iodide detector and RASCAL PRS-2 scaler (s/n 247). Radiation levels as low as 0.5 µR/h can be measured with an accuracy of +10% and -30%; 300 counts in a 30 second counting interval.

The energy and exposure rate responses of the detector system were determined using Cs^{134}, Co^{60} and Ra^{226} calibration sources. Accumulated counts in 30 seconds at a true exposure rate of 8 mR/h from Cs^{134} (0.605, 0.794 MeV), Co^{60} (1.17, 1.33 MeV), and Ra^{226} (.187 MeV plus others) were 967,000; 759,000 and 746,000 respectively. The Ra^{226} calibration factor was used to calculate exposure rates in this study. Therefore, the exposure rate can be overestimated by up to 30% as the energy of the radiation varies. The radiation fields around the Cs^{134} and Co^{60} sources were calibrated using a Farmer Dosimeter which itself has a calibration that is traceable to National Research Council standards. The Ra^{226} source was calibrated using an ion chamber and sources traceable to the United States National Bureau of Standards (NBS) (Ha84).

The exposure rate response of the detector was checked between 60 and 8000 μR/h using the Ra^{226} source. The calibration curve is plotted in Figure 4.1-1 and the non-linearity due to dead-time losses is apparent above 250 μR/h. The calibration curve detail below 250 μR/h is replotted in Figure 4.1-2. The calibration curve was forced to pass through the origin and the slope was calculated by least square fitting to the three data points shown. In the range 0 - 250 μR/h, the gamma exposure rate, ER μR/h, is calculated using the equation:

$$ER = CR \times 0.00159$$

where

CR = counts per 30 seconds
0.00159 = calibration factor in μR/h per count per 30 seconds

4.1.2 Beta Exposure Rate Surveys

Beta exposure rates were measured using an AEP 2153B monitor of Atomic Energy Canada Limited (AECL) design. The thin end window geiger tube has a removable steel window in front and can be used to measure beta exposure rates in the range 0 - 100 mR/h. In the presence of gamma radiation, the beta exposure rate is the difference between the two readings with the steel window open and closed.

Calibration of the monitor had previously been carried out using a P32 source. The absolute value of the activity of the source was determined by 4 pi beta counting. The accuracy of the beta dose rate was estimated to be ±50% (St84).

4.1.3 Beta-Gamma Exposure Rate Measurements

Extremity exposure rates for workers handling radioactive materials in the two facilities were measured using thermoluminescent dosimeters (TLDs). Various objects were placed in contact with the TLD for a recorded exposure interval. The lithium borate element No. 1 of the UD801 TLD manufactured by Panasonic was used for this study (see Figure 4.1-3). The elements of all plaques had been individually calibrated in the radiation field around a Co^{60} source. A Farmer Dosimeter (calibration traceable to the NRC) had been used to measure the exposure rate around the Co^{60} source.

The filtration over the lithium borate chip was determined after reference to the skin thickness of the hand reported in 'Report of the Task Group on Reference Man' (ICRP23). From the table in that report which lists the thickness of the epidermis for six main body site groups for males and females ages 15 - 89 years it was noted that the thickness for the fingers and fingertips was 220 and 370 μm respectively. The standard deviation was also reported to be 120 and 112 μm respectively. Based on a tissue density of 1.1 g/cm^3, these values were converted to equivalent mg/cm^2 and are shown in Figure 4.1-4. The filtration levels of 19 and 27 mg/cm^2 used in these measurements

are shown bracketing the thinnest epidermal region exposed when a radioactive item is picked up in the fingers.

It is appropriate to note at this point, that the most energetic alpha particles emitted by the thorium^{232} series, 8.79 MeV (Po^{216}), cannot penetrate the epidermis layer on the fingers and therefore are not considered an exposure hazard. The maximum range of an 8.79 MeV alpha particle in tissue can be calculated using equations from 'Introduction to Health Physics' (Ce76).

4.1.4 Alpha Contamination Surveys

Total and removable alpha contamination levels were determined by different methods, direct instrument measurement and wipe counting respectively. Total alpha contamination was measured on flat surfaces using a zinc sulphide scintillation detector (model 702 'Poppy' probe) and Thyac III ratemeter manufactured by Victoreen. The ratemeter has four ranges x1, x10, x100 and x1000 and the scale on the meter covers 0 to 800 counts per minute.

Calibration of the detector system was made using a standard source with a known alpha flux and traceable to the US NBS.

The counts per minute observed on the meter were converted to radioactivity densities by making the following calculation:

$$A = 2 \times C / (0.286 \times 1.27)$$

where

A = alpha activity density, dpm/100 cm^2
C = count rate observed on the meter, cpm

0.286 = detector efficiency relative to the 2 pi alpha flux emitted by the standard cpm per alpha/minute

2 = factor to convert 2 pi alpha flux to 4 pi alpha disintegration rate

1.27 = the ratio of the area (127 cm^2) of the detector face to 100 cm^2

The minimum sensitivity is 50 cpm using the lowest range. Since the area of the detector face is 127 cm^2, the minimum sensitivity corresponds to an alpha activity density of 280 dpm/100 cm^2.

Wipes taken to determine removable contamination were measured using a windowless gas flow proportional counter. The counter efficiency is checked regularly using the alpha calibration source indicated above. For the purposes of calculating the removable contamination on the surface wiped, it was assumed that only 10% of the activity stuck to the wipe and was counted. The background in the proportional counter is 0.5 cpm and the efficiency is 100% (not

including the geometry factor). The minimum sensitivity of the system for a 2 minute count is 1 dpm on the wipe and 10 dpm/100 cm^2 when the wipe has been dragged over 100 cm^2.

4.1.5 Thoron Gas Measurements

The concentration of thoron gas in air was measured using the two filter method described in the 'Manual on Radiological Safety in Uranium and Thorium Mines and Mills' (Ss43). A steel tube of inner length and diameter 50 cm by 5 cm respectively was fitted with 0.8 µm pore size membrane filters at each end. Air was drawn through at a rate of 10 +5% L/min and the flow was held constant by the use of a critical orifice. Between 7 and 24 hours after sample collection, the filters were alpha counted on the windowless gas flow proportional counter described above. The fraction of thoron decay atoms in the tube that deposit on the exit filter was calculated to be 0.664 using the appropriate equation from Appendix III (Ss43).

4.1.6 Thoron Daughter In Air Measurements

Thoron daughter concentrations in air were measured by drawing an air sample through a filter and measuring the alpha activity of the $Pb^{212}(Bi^{212})$ daughters in a windowless gas flow proportional counter. Total thoron daughter concentrations were measured in large air samples using an open face filter and sampling flow rates in the range from 10 to 30 L/min. Respirable thoron daughter concentrations in small air samples were measured using a flow rate of 1.7 L/min through a 10 mm nylon cyclone separator and a down-stream in-line filter. Membrane filters with 0.8 µm pore size were used in both cases.

The open face filter system consisted of the aluminum filter holder, a critical orifice to hold the flow constant at 10 or 14 L/min and a diaphragm air pump (GAST model DOL-101-AA). In a few cases, no critical orifice was used and the sampling flow rate varied from 33 to 27 L/min as the filter loaded up. During sample collection, the plane of the filter face was oriented perpendicular to the ground. If there was a significant flow of air, as in an exhaust duct, the plane of the filter face was also oriented parallel to the air flow.

The respirable size aerosol sampling system included a DuPont model 2500 personnel air sampling pump with self regulating flow and nylon cyclone separator and in-line filter described above.

The flow rates of both systems were calibrated using a bubble tube and stop watch. The uncertainty in the calibration of the flow rate is estimated to be no greater than $\pm$5%.

Alpha counting of the membrane filters was carried out on the windowless gas flow proportional counter described in Section 4.1.4. The minimum sensitivity of the method based on a 2 minute counting interval is 0.017 Bq.

4.1.7 Thorium^{232} Measurements

Thorium^{232} concentrations in air, soil and construction materials were measured by alpha counting or by neutron activation analysis.

Thorium^{232} concentrations in air were determined for total particulate and respirable particulate species. The air sampling methods have been described in the section on thoron daughter measurements and many of the same filters were used for both analyses. The amount of thorium^{232} on each filter was determined by the neutron activation method. After air sample collection, the filter was prepared for neutron irradiation with thorium standards prepared on filter papers. Counting of the activated samples and standards took place a few days after irradiation to allow short-lived interfering activities to decay.

The thermal neutron irradiation of Th^{232} produces Th^{233} which quickly decays (half-life 22.2 minutes) to Pa^{233}. Protoactinium^{233} has a half-life of 27 days and decays by the emission of a beta particle and numerous gamma rays including one at 312 keV. A hyperpure germanium detector was used to count the samples and standards using the 312 keV photopeak.

Thorium standards used in determining thorium on filter papers were prepared by pipetting commercially available atomic absorption standards onto blank filter papers. Sediment rock standards STSD-2 from the Canada Center for Mineral and Energy Technology (CANMET) and standard rock GSP-1 from the United States Geological Survey were used as standards for the determination of thorium in soils and roofing materials.

Thorium^{232} was also measured on some of the filters by alpha counting in the windowless gas flow proportional counter after the thoron daughter activity had decayed. The calculation of Th^{232} content was based upon the results of thoron retention and equilibrium measurements made with the alpha spectrometer and described in Section 4.1.9.

4.1.8 Air Velocity Measurements

Air velocity measurements in the labs, vents and air ducts were made using a Datametrics 100VT Air Flow Meter (s/n 2486) manufactured by Dresser Industries. The meter has two ranges, 0 - 600 feet per minute (FPM) and 600 - 6000 FPM and operates on the hot wire anemometer principle. The probe has a diameter of 9.5 mm and a length of 197 mm. It is connected to the control box by 2.3 m cable and can easily be manipulated into small openings.

The meter was calibrated on the upper range using an automobile speedometer which had been calibrated against Ontario highway kilometer markers. The lower range was calibrated using a uniformly rotating wheel and stopwatch. The calibration curves are shown in Figures 4.1-5 and 4.1-6 respectively.

4.1.9 Alpha Spectrometry - Age Of Thorium Stock Materials

The interval since refining of the thorium and the retained fraction of thoron gas in the thorium dioxide particles were measured by alpha spectrometry. A thin source was prepared, with no chemical treatment, and a surface barrier detector and multichannel analyser were used to determine the alpha spectrum.

A sample of thorium nitrate, similar to the stock material used at Facility A, was obtained by AECL Chalk River from May & Baker Canada Inc. and the alpha spectrum was measured. The alpha activity ratios were determined to be:

$$Th^{230}/Th^{232} = 0.18 \pm 0.04$$
$$Th^{228}/Th^{232} = 0.43 \pm 0.05$$

and the daughters of Th^{228} were in equilibrium (Jo84). The age of thorium since refinement and radium separation was estimated to be between 3 and 8 years. This estimate was based upon the variation of Th^{228}/Th^{232} activity ratio shown in Figure 4.1-7.

A sample of thorium dioxide, taken from stock material used at Facility B, was analysed in a similar manner. The alpha activity ratio was determined to be:

$$Th^{228}/Th^{232} = 0.60 \pm 0.045$$

Based on the activity ratio curve shown in Figure 4.1-7, the age of the thorium from Facility B is either 1.4 to 2.0 years or 10.5 to 13 years. Since the function is double valued it is impossible to determine which age range is correct based on only one measurement. The relative alpha count rates of the Th^{228} daughters are shown in Figure 4.1-8. Within the uncertainties indicated due to counting statistics all the Th^{228} daughters are in equilibrium.

It is significant to note that the Rn^{220} has been almost completely retained within the thorium dioxide particles. It is estimated that the escaping fraction of Rn^{220} is less than 2%.

4.2 Facility A Results

4.2.1 Inner Courtyard

In the past waste mantle material was burned in the inner courtyard shown in detail in Figure 4.2-3. The location of the courtyard within the plant is shown in Figure 4.2-1. Gamma exposure rates were measured at an elevation of one metre above the ground/asphalt using the NaI/ RASCAL system and the results are shown in Figure 4.2-3. In general the exposure rate above the asphalt surface is about 4 μR/h except near the center where elevated levels of 13 and 7.9 μR/h indicate the remains of thorium laden waste. The other two sections of the courtyard are bare soil and the corresponding exposure rates are higher, 6 to 7 μR/h due to lack of asphalt shielding.

Soil samples were taken at three locations in the courtyard and were analysed for Th^{232} by neutron activation analysis (NAA). Th^{232} in the sample taken around the sewer outlet was very high at 2330 parts per million (ppm) ±3%. The other two samples were measured at 155 ppm ±3.2% and 43 ppm ±4%, which are significantly above normal levels of 1 to 10 ppm.

4.2.2 The Test Lab

In the past, thousands of mantles were burned to test lifetime and other quality control measurements were carried out in this room. Measurements show that there still remains some residual radioactivity from these operations, see Figure 4.2-4.

Thoron daughter concentrations in air were measured on two different days using open face filters. The thoron daughter concentrations were calculated to be 1.7 ±0.29 and 0.8 ±0.22 Bq Pb^{212}/m^3 in air volumes of 1.35 and 1.2 m^3 respectively.

The same open face filters were analysed for Th^{232} by NAA. Th^{232} concentration in air was calculated to be <0.083 and <0.074 µg Th^{232}/m^3.

Gamma exposure rate measurements were made using the NaI/RASCAL system. The radiation level varied from background at 5 to 6 µR/h near the entrance and the drop test machine to a high of 12 µR/h near the mantle-burn test bench.

Removable contamination was measured by wipe test and alpha counting. The benches beside the entrance were not used for mantle related tests and had very little contamination <10 and 10 ±10 dpm/100 cm^2. However, significant removable contamination remains on the drop test and mantle-burn test benches.

4.2.3 Storage Room

Metal drums of thorium nitrate are no longer stored in the storage room and it retains no measurable trace of its former use.

Gamma radiation measurements were made with the NaI/RASCAL at one metre above the floor at the positions indicated in Figure 4.2-5. The results were indistinguishable from normal background and ranged from 4.7 to 6.6 µR/h. The thoron daughter concentration in air was measured on one occasion using an open face membrane filter and a 0.9 m^3 air volume. The thoron daughter concentration was calculated to be 0.4 ±0.2 Bq/m^3 and is within the range of normal background reported in (Tr142). The same filter was analysed for Th^{232} by NAA and the Th^{232} concentration in air was calculated to be <0.11 µg/m^3.

Removable alpha contamination was determined in the storage room and ranged from 10 to 60 dpm/100 cm^2.

4.2.4 (Pre 1970) Thorium Room

The dissolution of the thorium nitrate used to take place near the east wall of this room. Gamma radiation fields of 150 μR/h were measured at the wall one metre above the floor. The field decreased rapidly to 22 μR/h at a distance of 2 metres from the wall. The thoron daughter concentrations in air were calculated to be 4.6 Bq/m^3 (±9%) and 4.2 Bq/m^3 (±33%). Air volumes of 4.6 and 0.20 m^3 were pulled through an open face filter and the respirable air sampling system respectively. Th^{232} concentrations in air were determined to be <.04 and <.02 using the same filters (NAA). The removable alpha contamination levels in this area were found to range from <10 to 430 ppm/100 cm^2. Except one wipe, which indicated a contamination level of 9900 dpm/100 cm^2 on the wall one metre above the floor where the mixing took place.

On the other side of the wall, to the east of the (pre 1970) thorium room, the gamma fields are 34 μR/h at the wall over the drain. The open drain stretches for a distance of about 11 metres and gamma radiation above it is fairly constant at about 30 μR/h. Thoron daughter concentrations in air over the drain were measured on three occasions using open face filters. The results were 4.8, 1.1 and 0.57 Bq/m^3. Th^{232} was measured at <0.02 μg/m^3 on each filter.

4.2.5 First Floor Below Present Thorium Room

On the first floor below the present thorium room three different areas were designated, the east room, the south room and the machine shop (directly below the thorium room), see Figure 4.2-2.

The gamma radiation fields in the machine shop ranged from 14 to 45 μR/h with an average of 25 μR/h. The thoron daughter concentration in air was measured with an open face filter and respirable air sampler. The results were calculated to be 7.0 (±5%), 10.2 (±6%) and 6.4 (±5%) Bq/m^3. The Th^{232} on each of these filters indicated <.04 Bq/m^3. Five floor wipes indicated 30 to 90 dpm/100 cm^2 removable alpha contamination. Higher levels up to 1000 dpm/100 cm^2 were found on top of cabinets and ductwork using 5 additional wipes.

The gamma radiation fields in the east room were a maximum of 14 μR/h at the wall below the thorium room and decreased to normal background within a few metres. An 8.3 m^3 air sample was taken with an open face filter and the thoron daughter concentration in air was calculated to be 1.4 Bq/m^3 ±7%. The Th^{232} concentration in air was <.02 μg/m^3.

A 6.2 m^3 air sample was used to determine the thoron daughter concentration in air in the south room. It was calculated to be 0.75 Bq/m^3 ±5%. The Th^{232} concentration was 0.006 (±28%) Bq/m^3.

4.2.6 Present Thorium Room

Thoron daughter concentrations in air were measured on 7 occasions using open face filters. The results ranged from 5.8 to 34 Bq/m^3 with an average of 16.9 Bq/m^3. The highest values were observed in the winter at 23 and 34 Bq/m^3 and the lowest values, 9.3 and 15.7 Bq/m^3 after the new thorium room exhaust fans had been installed and operated in the fall.

Th^{232} concentrations in air were also measured using 6 of the open face filters; results ranged from <0.1 to 0.23 µg/m^3 with an average of 0.13 µg/m^3. Once again the highest values were observed in the winter. The results of the respirable air samples taken in the fall were <0.113 and <0.06 µg/m^3.

The average gamma radiation exposure rate in the thorium room is 50 µR/h. In the corner where the thorium nitrate used to be dissolved the exposure rate increases to 200 µR/h.

The removable alpha contamination was measured using 10 wipes and the results ranged from 90 to 1700 dpm/100 cm^2. The five floor wipes indicated 90 to 870 dpm/100 cm^2. The remaining 5 wipes of shelves and the oven top etc. were generally higher. The 1700 dpm/100 cm^2 value was found on the lower shelf of the bench where thorium solutions were prepared.

4.2.7 Mantle Manufacturing

In the mantle manufacturing area the thoron daughter concentrations in air were measured in the winter using open face filters and they were calculated to be 6.3 Bq/m^3 (±13%) and 7.1 Bq/m^3 (±11%). Two other measurements were made in the fall after the new exhaust system had been installed in the thorium room. The thoron daughter concentrations in air were calculated to be 5.8 Bq/m^3 (±10%) using the radon tube and 8.3 Bq/m^3 (±20%) using the cyclone separator and personal sampler.

Th^{232} was measured by NAA on two of the same air filters and the concentration in air was calculated to be <0.082 µg/m^3 using the radon tube and <0.097 µg/m^3 using the cyclone separator and the personal air sampler.

Gamma radiation fields were found to average 15 µR/h except near wooden trays etc. that had been used to stack thorium laden materials. In these areas the exposure rate increased to 50 µR/h.

4.2.8 Lunch Room And Adjacent Areas

The results of the gamma survey of the lunch room indicate that natural background radiation fields of 4 to 6 µR/h prevail throughout. In the adjacent manufacturing areas the gamma radiation fields at the walkway beside the mantle manufacturing area were 9.7 µR/h but

decreased to natural background a few metres from the walkway.

The thoron daughter concentrations in air were measured in the lunch room and adjacent manufacturing area using open face filters. There was *very little* difference in the results in winter and fall; the average of the five values was 4.7 Bq/m^3 with a standard deviation of 0.63 Bq/m^3.

4.2.9 Ladies Change Room

The gamma radiation fields were generally 8 μR/h and increased to 12 μR/h at the wall adjacent to the thorium room.

In the ladies change room eight measurements of thoron daughter concentrations averaged 11.5 Bq/m^3 with a standard deviation of 3.9 Bq/m^3. These represent five open face filter samples and three samples of respirable size particles.

Th^{232} concentrations in air were measured three times with open face filters, <0.125, <0.08 and <0.10 $\mu g/m^3$, and once with the cyclone separator, <.147 $\mu g/m^3$.

The removable alpha contamination was measured using 5 wipes and the results ranged from 10 to 150 dpm/100 cm^2.

4.2.10 Roof Exhaust - Environmental

The air flow rates out of the thorium room exhaust stacks were measured with the hot wire flow meter. The air speeds, duct sizes, etc. are given in Table 4.2-1. The volume of the thorium room is 465 m^3 and the air change rate varies from 18 to 23 per hour when the room is occupied and the cabinet exhausts are on.

The thoron daughter concentrations in roof exhaust air was measured at each of the dryer, lacquer, and ammonia cabinet exhausts and the old and new exhaust systems from the thorium room itself. The concentrations ranged from 9.0 to 13.2 Bq/m^3 and were very similar to thorium room values. This is to be expected since the exhausts are unfiltered.

4.2.11 Contact Beta-Gamma Exposure Rate

The contact beta-gamma exposure rates were measured with TLDs on two types of lantern mantles. The results are shown in Table 4.2-2. In a 2000 hour working year the maximum dose to the fingers could be as high as 9600 mRad.

4.3 Facility B Results

Thoron daughter concentrations in air were measured in the mixing, coating and storage rooms and all results were <1 Bq/m^3.

4.3.1 ('65-'78) Coating Room

The gamma radiation fields in this area were as high as 15 μR/h in one spot but generally were at the background level of 5 μR/h.

Total alpha contamination levels on the floor were less than 280 dpm/100 cm^2 in all areas including where the varathane floor covering had been worn away to expose the bare concrete. Removable alpha contamination was non-detectable on all four wipes and therefore is less than 10 dpm/100 cm^2.

4.3.2 Waste Storage

Gamma fields in the waste storage area were generally at the natural background level except in the vicinity of the 45 gallon drums of waste. Contact radiation readings varied up to 0.5 mR/h.

Th^{232} concentration in air was below the detection limits on 3 different days at <.06, <.06 and <.16 μg/m^3. Removable alpha contamination was <50 dpm/100 cm^2.

4.3.3 Electrode Assembly

Gamma radiation fields were at the natural background level throughout the area at about 5 μR/h. At the wall beside the coating room the gamma field was as high as 20 μR/h.

The total alpha contamination was found to be less than 280 dpm/100 cm^2 throughout the electrode assembly, electrode press and furnace areas. Removable alpha contamination on the floor areas outside the coating room was 70 dpm/100 cm^2 and less except on the table where electrodes were welded. There the removable alpha contamination was 180 dpm/100 cm^2.

Th^{232} concentrations in air were measured at .124 and .038 μg/m^3 with open face filters and .036 μg/m^3 with the cyclone separator.

4.3.4 Coating Room

Gamma radiation fields in the coating room averaged 37 μR/h throughout the room and were a maximum of 50 μR/h at the storage rollers for the thorium stock solutions.

In the coating room total alpha contamination levels were measured on the floor in the range of 280 to 840 dpm/100 cm^2. On the work

bench the levels were much higher at 5500 to 7000 dpm/100 cm^2. Removable alpha contamination was only 50 dpm/100 cm^2 on the same bench top where the total alpha was 7000 dpm/100 cm^2. The highest level of removable contamination was 6000 dpm/100 cm^2 in the thorium storage cabinet. Of the other 7 wipes taken on floors and benches the highest was 890 dpm/100 cm^2.

During the time that subject B10 was coating electrodes the Th^{232} concentrations in air were measured at 3.8, 3.7, 3.0 and 1.96 $\mu g/m^3$ with the cyclone separator. While B3 was doing the same work Th^{232} concentrations in air were measured at 2.0 and 0.92 $\mu g/m^3$.

4.3.5 Coating Room Exhaust - Environmental

Gamma radiation readings were made around the exhaust fan, filter and ductwork which removes the fumehood air from the coating room. A radiation reading of 115 $\mu R/h$ at contact with the filter box indicates the presence of thorium. Along the ductwork, radiation fields of 32 $\mu R/h$ were found above the duct and 80 $\mu R/h$ was found below, indicating that thorium powder was lying inside the duct itself.

Thoron daughter concentrations were measured in the exhaust air from the coating room and were calculated to be less than 1 Bq/m^3.

Th^{232} concentrations in exhaust air were calculated to be 2.6, 1.3 and 9.4 $\mu g/m^3$ even though there is an inline filter.

Roofing gravel was collected at a distance of 4 metres from the coating room exhaust stack and analysed for Th^{232}. The thorium concentration was measured at 75, 74, 108 and 99 ppm to the north, south, east and west respectively. These levels are much higher than the 2.0 ppm that was measured in a roofing gravel sample taken from a section of roof about 50 metres from the vent.

4.3.6 Mixing Room

In the mixing room the total alpha contamination is 3600 dpm/100 cm^2 in front of the storage rollers and 660 to 1400 dpm/100 cm^2 in between the glove box and ball mill. The remaining floor and bench areas of the mixing room measured less than 280 dpm/100 cm^2. There is very little removable contamination on the floors and benches throughout the lab; maximum is 260 dpm/100 cm^2. Significant removable contamination was found on the plastic bottles used to store the thorium solutions, 1240 dpm/100 cm^2; under the storage rollers, 9480 dpm/100 cm^2; under the ball mill, 3290 dpm/100 cm^2; on the ball mill, 7320 dpm/100 cm^2 and in the drawer used to store thorium transfer and measuring containers.

Gamma exposure rates throughout the room average 55 $\mu R/h$ except at the storage rollers, 110 $\mu R/h$ and at the glove box, 800 $\mu R/h$.

Personal air sampling with the cyclone separator was carried out while subject B7 prepared thorium dioxide solutions. On one occasion a Th^{232} concentration of 13.8 $\mu g/m^3$ was measured over a period of 1.5 hours and 0.96 $\mu g/m^3$ was measured over 7.5 hours. While non thorium work is carried on in the mixing room the Th^{232} concentrations in air are quite low, e.g. 0.19 and <0.09 $\mu g/m^3$.

Immediately to the east of the mixing room is a small office with two desks. The exposure rates at the desks are 33 and 17 $\mu R/h$ respectively and at the wall common to the mixing room it is 52 $\mu R/h$. These fields originate from the thorium dioxide stock stored in the glove box. On the south wall of the mixing room there is an exposure rate of 25 $\mu R/h$ from the thorium stored in the bottles on the rollers.

4.3.7 Mixing Room Exhaust - Environmental

Soil samples were taken from directly under the exhaust duct and from an area 10 metres to the north. The Th^{232} concentrations were found to be 2.9 ppm (±7.1%) and 2.3 ppm (±11%) respectively.

Exhaust air was sampled using open face filters and they were analysed for Th^{232}. Dayshift Th^{232} concentrations in the exhaust air were calculated to be 0.03 (±10%), 0.049 (±4%), <0.02 and <0.0079 $\mu g/m^3$. During the nightshift the concentrations were <0.005 $\mu g/m^3$.

4.3.8 Contact Beta-Gamma Exposure Rates

The contact beta-gamma exposure rates were measured with TLDs on pieces of thorium metal and finished electrodes (Table 4.2-3). The results show that the exposure rates average 30 and 17.3 mR/h with 19 mg/cm^2 filtration and 19 mg/cm^2 filtration plus a cotton glove respectively. At the 27 mg/cm^2 filtration level the exposure rate is no different with or without the glove within the accuracy of these measurements. Although the metal is not handled very often, the wisdom of using tweezers and not relying on gloves is apparent.

The contact exposure rate from electrodes is 0.63 mR/h and less, as would be expected, because of the small amount of thorium on them.

5.0 PERSONNEL MONITORING

Bioassay measurements for thorium and it's daughter products were made on volunteers from both facilities. The measurements included in vivo determinations by whole body counting, lung counting and thoron in breath measurements. Also, 24 hour urine and fecal samples were collected and were analysed for thorium and radium. Volunteers were transported to in vivo counting facilities at the Radiation Protection Bureau (RPB) Health and Welfare Canada in Ottawa and the Atomic Energy of Canada Limited Research Company (AECL-RC) Laboratories at Chalk River. Urine and fecal samples were collected at the volunteers' homes and were sent to AECL-RC for analysis. The

subjects with detectable lung burdens are also those subjects with long work histories in areas of highest potential thorium uptake.

5.1 Methods And Calibrations

5.1.1 Whole Body Counting

Whole body burden measurements were made of subjects from Facilities A and B in the shadow shield at (AECL-RC) laboratories at Chalk River. The shadow shield is located in the Low Background Building No. 560 and the detector is a 29 cm diameter by 10 cm thick sodium iodide crystal located in the top of the shield. A complete description of the equipment and the method is given in (Pe83).

Before the measurement, subjects were required to shower and wash their hair and they were provided with a clean set of disposable clothes. For the first part of each measurement the subject assumed a prone position on the shadow shield stretcher. The stretcher is powered by a worm gear and transports the subject under the detector at a uniform speed. For the second part of the measurement the subject assumes a supine position and is transported back to the starting position.

Sensitivity tests have been made using a bottle phantom and they have shown that variations in the redionuclide distribution in the phantom (in head, thorax, trunk, etc.) result in less than a 10% change in the count rate per becquerel of body burden.

The gamma ray that was used to determine body burden in this case was the 2.6 MeV radiation from T1208. The results are quoted on the basis of Th^{232} activity assuming that secular equilibrium has been established, that no thoron gas escapes in either the subject or the phantom and taking into account the 36% branching ratio for T1208. The calibration factor used here was based on a model given in (Pe83).

5.1.2 Lung Counting

Lung/organ burden measurements were made on the subjects from Facilities A and B at AECL-RC labs at Chalk River and at the RPB in Ottawa.

At Chalk River, the lung burden measurement equipment consists of two phoswich detectors, amplifiers and multichannel analyser and is located in Building No. 560. Each detector has a diameter of 12.7 cm, 0.1 cm thick front crystal of NaI(Tl) and 5 cm thick back crystal of CsI(Tl). The female subjects were measured in a prone position with hands under the head and the phoswich detectors were positioned over the lungs above the back. The male subjects were measured in a supine position with hands at the side and the phoswich detectors were positioned over the chest. A net count for each subject was calculated by subtracting a background count from the subjects gross count. The background value to be subtracted was established by measuring

a matched subject with no lung burden. The calibration factor applied to the net counts was determined by measurement of the Lawrence Livermore Phantom (from RPB) with thorium loaded lungs. The radiations detected by this method are mainly the 75, 77 and 87 keV x-rays from Pb^{212} (Ko81). The results are quoted on the basis of Th^{232} activity assuming that secular equilibrium has been established and that no thoron gas escapes in either the subject or the phantom. A complete description of the equipment and the method is given in (Jo76).

Organ burdens of the subjects from both facilities A and B were measured on one occasion at the Radiation Protection Bureau in Ottawa. Two planar hyperpure germanium detectors were used to make a total of four measurements on each subject. Alternately, the two detector set was located 1 cm above the dorsal surface of the subject over the right lung, left lung, liver and knee. Each of the resulting gamma spectra was searched for the presence of photopeaks at 75, 77 and 87 keV which would indicate the presence of Pb^{212}. The net area of the combined photopeaks was determined by subtracting the background beneath it. This background was calculated from the subject's own spectrum by assuming a straight line between the background levels on each side of the photopeaks.

The detector system was calibrated for right and left lung measurements using the Lawrence Livermore Phantom and the lung set loaded with natural thorium. Liver and knee joint calibrations were not possible because the corresponding thorium loaded organs are not yet available. For the purposes of calculating organ burdens it was assumed that all of the thoron gas was retained in both the phantom's and the subjects' lungs and that secular equilibrium had been established throughout the Th^{232} chain down to Pb^{212}.

5.1.3 Thoron In Breath

The thoron in breath measurements were made on each of the subjects at AECL-RC using the following method. The subject was required to lie in a restful position for 30 minutes and breath into the mouthpiece of the apparatus. The air was directed into a 37.7 litre metal cylinder in which essentially all of the thoron gas decayed. The Pb^{212} was collected on a small disc which was held at 4000 volts relative to the container shell. After collection the samples were held for 170 minutes to allow the decay of interfering $radon^{222}$ daughters. The Pb^{212}-Bi^{212} was then counted on a zinc sulphide scintillation detector during the 170 - 1010 minute interval after sample collection.

About 50 (uncontaminated) controls have been counted at AECL-RC and it has been concluded that 20 - 30 counts in the 170 to 1010 minute counting interval represents normal background. A count above 40 likely indicates a lung burden. The exhalation rate of thoron (Rn^{220}) in mBq per minute by the subject can be calculated by multiplying the counts in the 170 to 1010 minute interval by a factor of 0.002 mBq per minute per count.

5.1.4 Urinalysis

Urine samples were collected from the subjects during weekends while they were away from the work environment. Each voiding throughout a 30 hour period was collected in a separate container. The samples which most closely approximated voidings from a continuous 24 hour period were combined and sent for thorium, Ra^{224} and Ra^{228} analysis.

The thorium analysis follows the gross (actinide) alpha method described in (Kr83-1). The Th^{232} and Th^{228} which were precipitated from each sample were alpha counted with a detection limit of about 0.1 mBq. The Ra^{224} analysis follows the calcium phosphate precipitation method described in (Kr83). The separated sample is immediately alpha counted with an estimated detection limit of 0.2 mBq. The Ra^{228} measurement is carried out on the samples prepared for Ra^{224} analysis after a 90 day period has passed to allow the ingrowth of Th^{228} and the decay of Ra^{224}. The alpha count of the Th^{228} results in a detection limit of about 1 mBq per sample. Above the detection limit the major source of uncertainty is due to variations in the chemical recovery from one urine sample to another as the pH or solid fraction varies. The recovery is typically 75% but can vary by plus or minus 25%.

5.1.5 Fecal Analysis

Fecal samples were collected from the subjects at their homes and stools which most closely approximated 24 hour samples were combined and sent for analysis.

The samples were chemically digested and then the analytical techniques described for the urinalysis were used for the fecal samples also.

5.1.6 Hair and Nail Analysis

Samples of hair and finger and toe nails were collected from each of the subjects. The samples were measured for Th^{232} content by neutron activation analysis using the method described in Section 4.1.7. The subjects were asked to remove all nail polish and thoroughly wash the hair before donating the samples. However, the samples were not cleaned in any other way.

5.2 Facility A Results

5.2.1 Whole Body Counting

Nine subjects from Facility A were measured in the Whole Body counter at AECL-RC in March 1984. Three subjects were taken back 3 months later in June for repeat measurements. Measurements were not repeated on the other six subjects because their lung and body burdens were below the detection limits of the systems used and there

was negligible potential for significant thorium uptake in the interim. The results of the measurements are shown in Table 5.2-1. Only subject A6 had a measurable burden and within the statistical accuracy of the measurement there was no difference between the two results.

5.2.2 Lung Counting

The same nine subjects from Facility A were measured in the lung counter at AECL-RC in March 1984 and three were repeated in June. Only subject A6 had a measurable burden and within the statistical accuracy of the measurement there was no difference between the two results. The results are shown in Figure 5.2-1.

The nine subjects from Facility A were also measured at RPB in March 1984. Subject A5 was the only one identified as having a measurable burden. However, it was concluded that it was likely due to external contamination. The burdens of the other 8 subjects were below the detection limits of the system.

5.2.3 Thoron in Breath

Nine subjects from Facility A were measured for thoron in breath at AECL-RC in March 1984 and three were retested in June. The results shown in Table 5.2-2 are the counts in the 170 to 1010 minute interval after sample collection and the calculated thoron exhalation rate in mBq/minute at the mouth. Only three subjects were exhaling thoron at a rate that was considered greater than normal in March. Repeat measurements on these three indicated that two of them, A6 and A7 had no change in thoron exhalation rate whereas, subject A9 which had been on the borderline in March was likely exhaling thoron at the normal rate of up to 80 mBq Rn^{220} per minute.

5.2.4 Urinalysis

The March and August urinalysis results are given in Tables 5.2-3 and 5.2-4 and are summarized in Table 5.2-1. ICRP-23 reports that the normal rate of thorium excretion in urine is 0.1 µg/day (0.4 mBq per day). Also, ICRP-23 reports that Ra^{226} is normally excreted in urine at 0.08 pCi/day (3 mBq/day). The results of the March urinalysis tests indicated Th^{232} concentrations in urine significantly above that of the normal population. However, this was not supported by the results of the second set of tests in August which were much lower. Also, there was no correlation between the two sets of results. The Ra^{224} elimination rates are similar in the normal population. The Ra^{228} results while large in some cases are not correlated with lung burdens nor thoron in breath results and they reflect normal excretion rates of radium. No Ra^{224} and Ra^{228} results were obtained from the March samples because contaminated resin interfered with the measurements.

5.2.5 Fecal Analysis

The results of the March fecal analysis are shown in Table 5.2-5. ICRP-23 reports that normally thorium is excreted in feces at 2.9 µg/day or 12 mBq/day. Also, Ra^{226} is normally excreted in feces at 2.2 pCi/day or 81 mBq/day. Since the measured excretion rates are those expected from normal subjects it was decided that the second set of fecal testing not be made.

5.2.6 Hair And Nail Analysis

The results of the neutron activation analysis of hair and nails are given in Table 5.2-6. The thorium in finger and toe nails was not correlated with the lung burden and thoron in breath results. The Th^{232} in the hair of A6 suggests a relationship with thorium lung burden however this is not supported by the result for subject A7.

5.3 Facility B Results

5.3.1 Whole Body Counting

Ten subjects from Facility B were measured in the Whole Body Counter at AECL-RC in March 1984. Six subjects were retested five months later in August. Subjects B2, B3, B6 and B7 were remeasured because the results of the March lung burden and/or thoron in breath measurements indicated possible thorium burdens. Subject B8 had just begun work in the coating room in January 1984 and it was considered prudent to retest this subject. Subject B10 had a long history of potential thorium exposure but had missed the March measurements because she had been on maternity leave. The results of both sets of whole body measurements revealed no subjects with body burdens above the detection limit (Table 5.3-1).

5.3.2 Lung Counting

Ten subjects from Facility B were measured in the Lung Counter at AECL-RC in March 1984 and six were measured in August. Only two subjects had measurable burdens; within statistical uncertainty there was no change in B3's burden and subject B10 had only been measured once (Table 5.3-1).

The ten subjects from Facility B were also measured at RPB in March in 1984. Subject B3 was reported to have 5 $\pm$15 mg Th^{232} (20 $\pm$ 60 Bq) in each lung. This is in some agreement with the result reported by AECL-RC. The other nine subjects (excluding B10) were reported to have lung burdens below the detection limits of the measurement technique.

5.3.3 Thoron In Breath

The results of the thoron in breath measurements on the subjects from Facility B are given in Table 5.3-2. The counts on each filter between 170 and 1010 minutes after the end of sample collection have been converted to thoron exhalation rates at the mouth. Between March and August subject B2 apparently dropped from 167 to 30 Bq/min at a statistically significant level. The results for three other subjects B3, B6 and B7 remained unchanged and the results for B8 increased slightly between March and August.

5.3.4 Urinalysis

The urinalysis results are reported in Tables 5.3-3 and 5.3-4 and the adjustment for excretion period has been made. The results are summarized in Table 5.3-1 and it is apparent that the excretion rates are similar to those observed in the normal population where rates can vary by an order of magnitude.

5.3.5 Fecal Analysis

The fecal analysis results did not have to be adjusted since they already represented outputs for 24 hours. The results of the fecal analysis are shown in Table 5.3-5 and also are not significantly different than the normal. Therefore, the second set of fecal analysis was cancelled for Facility B subjects also.

5.3.6 Hair and Nails Analysis

The results for the hair and nail measurements are reported in Table 5.3-6. The results are random and not correlated with lung burdens nor thoron in breath measurements.

6.0 SOLUBILITY IN SIMULATED LUNG FLUID

The solubility in simulated lung fluid, SLF, tests were carried out at AECL-RC in Chalk River and included the measurement of thorium, radium-224 and uranium.

The simulants and methods used have been described by Eidson and Griffith in the Health Physics Journal (Ei84). The composition of the simulated lung fluid is given in Table 6-1 and the method was similar to the batch/filter method described by Eidson. A weighed portion of material was added to the SLF, the pH was adjusted to 7.3 and the container was sealed with a CO_2 atmosphere above the solvent. The sample was agitated in a shaker throughout the experiment and remained at room temperature. Five days out of seven the SLF was centrifuged and filtered to separate the dissolved and undissolved thorium. Fresh SLF was added to the undissolved thorium and the sample was prepared again as described above. The supernate was

chemically separated and the uranium, Ra^{224} and thorium (gross alpha) concentrations in SLF were measured by alpha counting.

A one gram sample (3760 Bq of Th^{232} assuming pure ThO_2) of thorium dioxide from Facility B was subjected to the solubility test described above. The concentrations of soluble thorium, uranium and Ra^{224} in SLF were measured on days 1, 3, 10 and 30 and the results are given in Table 6-2.

Based upon a SLF batch size of 300 ml, and a starting activity of 3760 Bq thorium the amount dissolved per day in Bq/day and as a fraction of the undissolved thorium have been calculated and the results are shown in Table 6-3. The first day results are higher than the other three and represent a slightly greater dissolution rate which is insignificant with respect to the following analysis.

The fractional dissolution rates of thorium for days 3, 10 and 30 are not different at a statistically significant level and were used to calculate the dissolution half time of 690 years - definitely a Class Y material.

The uranium contamination in the thorium dioxide is also class Y based upon the dissolution rates given in Table 6-2.

The Ra^{224}/Th^{232} activity ratio in the Facility B thorium dioxide was measured at 0.60 ± .045 (section 4.1.9). Therefore, the undissolved Ra^{224} activity at the beginning of the study was 3760 x 0.60 = 2256 Bq (±7.5%). The dissolution rate was fitted by a function which was the sum of two exponentials; 85% of the radium had a dissolution constant of 9.0×10^{-5} per day (Y class) and the remaining 15% has a dissolution constant of 0.045 per day; a W class material.

A thorium nitrate sample from Facility A was also tested for solubility in simulated lung fluid. As soon as the one gram of material was added to the 300 ml of SLF the nitrate dissolved. Almost immediately it formed an insoluble complex (perhaps phosphate) with the fluid. The material although not initially a class Y material had been transformed into one. The experiment was therefore not continued.

7.0 EVALUATION OF RADIOLOGICAL SIGNIFICANCE AND RECOMMENDATIONS

During the course of the measurements at the two facilities it became apparent that some aspects of the radiological significance of the findings were distinctly different at the two plants. At Facility A the risk to non Atomic Radiation Workers was primarily due to thoron daughters in air while gamma radiation and Th^{232} in air were less important. On the other hand, at Facility B the inhalation of Th^{232} is the significant exposure pathway.

The significance of the results of the bioassay measurements at both plants were similar. In vitro measurements of radioactivity in urine, feces, hair and nails were not good indicators of lung or body burdens.

Lung burdens were identified in subjects from both plants by gamma spectrometry and thoron in breath techniques.

A6 was the only subject from Facility A who had a detectable lung burden by whole body or lung counting. The statistical uncertainty in each measurement is so large that none of the results can be said to be different than any of the others. Therefore, the average burden and the observed standard deviation among the results were calculated to be 37 ± 13 Bq Th^{232}. Subject A6 had worked with thorium at Facility A from 1955 to 1983. If a chronic exposure for a period of 28 years is assumed, then the lung and lymph burden resulting from the inhalation of a Y Class material was calculated to be 130 + 96 = 226 units for one unit per day intake using Johnson's ICRP lung model tables (Jo83). Therefore, a lung and lymph burden of 37 ± 13 Bq would result from a daily intake of 37/226 = 0.164 ± 0.051 Bq/day. Based on the Th^{232} dose factor for inhalation of Y Class material (3.7×10^{-3} Sv/Bq) (Jo83) the annual increment in the committed effective dose equivalent for each of the 28 years would have been:

$$0.164 \times 365 \times 3.7 \times 10^{-3} = 0.022 \pm .008 \text{ Sv.}$$

There would be an additional dose from the inhalation of Th^{228} which was measured at 0.43 ± .05 relative to the activity of Th^{232}. The annual increment in the committed effective dose equivalent from the Th^{228}, dose factor of 1.1×10^{-4} Sv/Bq (Jo83), would have been:

$$0.164 \times 0.43 \times 365 \times 1.1 \times 10^{-4} = 0.0028 \pm .001 \text{ Sv.}$$

Other daughters of Th^{232} would have been inhaled also but the doses would have been insignificant because their dose factors are much lower than those for Th^{232} and Th^{228}. In total, assuming a chronic exposure over 28 years, the increment in the committed effective dose equivalent rate to subject A6 from the inhalation of thorium nitrate would have been 0.025 ± .0075 Sv per year.

Similar dose calculations were made for the subjects B3 and B10 using their respective lung burdens and years of exposure. The annual increments in the committed effective dose equivalent rates were calculated to be 0.017 ± 0.011 Sv per year for 11 years and 0.025 ± .018 Sv/y for 7 years respectively. The doses to subjects A6, B3 and B10 represent 50%, 34% and 50% respectively of the limit of 0.05 Sv/y for Atomic Radiation Workers. With appropriate modifications to equipment and practices as described below, future doses can be reduced to much lower levels.

The results of the thoron in breath measurements on all three subjects who had lung burdens detected by lung and whole body counting were above the level for normal uncontaminated subjects. In addition, the thoron in breath technique identified two other subjects, A7 and B6, who likely had lung burdens below the detection limits of the other methods. The calculation of lung burden using thoron in breath results is very sensitive to the rate at which thoron gas escapes from the thorium particles in the lung and into the mouth. It is therefore necessary to determine the actual particle size distribution

of the thorium in the lung. The rates at which thoron escapes from the mouths of subjects A6, B3 and B10 have been calculated relative to the measured lung burden as 1.0 ± .5%, 1.7 ± 1.8% and 2.8 ± 2.3% respectively. Thoron in breath measurements are the most sensitive indicator of lung burdens but cannot be used to calculate lung burdens with presently known data.

The fractions of the annual effective dose limit (5 mSv/y) received by non Atomic Radiation Workers occupying each room in Facility A for 2000 hours per year have been calculated and are shown in Figure 7-1. The calculations have been based upon the inequality given on page 54 of ICRP 30 Part 1 and the following limits were used: 0.88 $\mu g/m^3$ Th^{232} and daughters; 33 Bq/m^3 thoron daughters (ICRP 32) and 2.5 $\mu Sv/h$ gamma radiation. In the four rooms with the largest fraction of the dose limit, thoron daughter concentration in air was the predominant contributor in each case.

Environmental pathway analyses were made using existing metabolic and dosimetric models to determine the maximum acceptable concentration of Th^{232} in soil that could be allowed to safely pass out of control of the licencee. Calculations were made for all pathways indicated in Figure 7-2 and the critical pathway was found to be the soil-vegetation-man pathway with the critical group, adults. It was assumed that the land was being used by a group of adults who consumed fruits and vegetables grown on the thorium laden soil. For the purposes of these calculations, it was assumed that each adult consumed 255 kg per year (Ss57) of fruit and vegetables. Transfer factors for Th^{232} and daughters from soil to vegetation were taken from reference Ss57 and the corresponding dose factors were taken from Jo83, see Table 7-1.

It was calculated that under the above circumstances a thorium in soil concentration of 0.25 Bq Th^{232}/g soil, (62 μg Th^{232}/g soil), would result in a committed effective dose equivalent rate of 5 mSv per year to each member of the critical group. Therefore, 62 $\mu g/g$ should be considered the upper limit for soil to be released from the licencee's control into the public domain.

Thorium processing was discontinued at Facility A, however, in the future workers will carry on other tasks in the areas indicated in Figure 7-1. In some areas remedial action is necessary to ensure that no member of the general public receive a radiation dose exceeding 5 mSv in any given year. Also, some simple cleanup operations have been identified which will reduce actual or potential doses in keeping with the ALARA principle, "as low as reasonably achievable, taking into consideration other economic and sociological factors."

The walls and floor near the mixing area in the pre-1970 thorium room should be decontaminated and the walls and floors sealed (with paint and varathane for example). When the property is sold or renovated these sections of wall and floor should be disposed of in an approved dump site.

No action is required in the east room, south room, lunch room and adjacent manufacturing areas on the second floor.

The machine room, thorium room and mantle manufacturing area on the second floor require remedial action in two stages. At the next convenient shutdown these areas should be decontaminated and the wooden benches disposed of to an approved dump site. The walls and floors should be sealed with paint and varathane to reduce the escape of thoron gas into the room air. In the longer term when major renovations are next carried out or the property is sold, the thorium impregnated walls and floors should be taken up and disposed of to an approved dump site.

The ladies change room should have continuous ventilation to reduce the thoron daughter concentration in air. The exhaust fans were shut down during the time when all the air sampling was carried out.

The inner courtyard requires no remedial action at this time. However, when renovations are carried out or upon the sale of the property, the waste material should be removed and transported to an approved dump site.

The test lab requires only cleanup of the removable contamination that remains on the drop test equipment and the burn test bench.

The storage room requires no action as no traces of the previous uses remain.

The following changes are recommended at Facility B. In the mixing room:

- The glove box should be increased in size so that all of the equipment for thorium preparation can be kept inside it. This will eliminate the potential for the spread of thorium contamination when the equipment is put into storage in the drawers.

- The procedure for preparing the thorium mixture should be modified to allow the addition of liquid to the thorium powder in the glove box. Then the transfer to the ball mill will involve a wet transfer rather than the current dry transfer. There would be a reduction in the thorium particulate in air concentrations to which the workers would be exposed.

- All thorium laden waste including wet rags and liquids should be disposed of into the radwaste collection system. This may require the use of liquid absorbent such as selite but would likely be cheaper than installing a holding tank and analysing the liquid before dumping it into the sewer.

- Gloves that are worn during sample preparations and become contaminated should not be put into the glove box gloves.

In the coating room a number of changes are recommended with reference to equipment and procedures for reducing thorium inhalation.

- Operator habits regarding smoking should be improved. The smoking ban in the coating room should be maintained and a special effort should be made to impress upon the workers that improved hygiene habits can significantly reduce thorium uptakes. Specifically, cigarettes should not be handled with gloves (or hands) contaminated with thorium powder.

- A glove box should be installed in which all stages of the coating operations can be carried out. It should be large enough to include the ultrasonic bath, the evacuation chamber and a small garbage container.

- Waste removal from the glove box should take place as follows; the plastic bag containing waste in the glove box should be closed and tied. At removal from the glove box it should be put into a clean bag and again tied.

The exhaust system for the coating room and the procedures associated with it should be modified as follows;

- When filter changes are made, a plastic bag should be positioned so that the filter can be transferred directly into it to minimize the spillage of dust. Any dust that is spilled should be cleaned up. A vacuum cleaner with an absolute filter is appropriate for this task.

- The thorium powder is very fine and an absolute filter should be used in the exhaust duct. To extend the lifetime of the absolute filter it is prudent to install a prefilter to catch dust and large particulates.

- The horizontal section of the exhaust piping is loaded with thorium dust and should be cleaned out. When this cleaning is carried out it would be advisable for the workers to wear dust masks and wet the thorium powder where possible.

- Workers in the mixing and coating room under present conditions must be classified as Atomic Radiation Workers. If the annual effective dose equivalent for these workers cannot be reduced below 30% of the limit, individual personal air sampling should be instituted as per the recommendations regarding Working Condition 'A' in Basic Requirements for Personnel Monitoring (Ss14). However, it is highly likely that the installation of and proper use of a glove box will reduce the dose rate and eliminate the need for personal monitoring. Semiannual lung burden and thoron in breath measurements should be established as routine until intakes have been shown to be effectively reduced.

8.0 CONCLUSIONS

Solubility in simulated lung fluid measurements on samples of the thorium dioxide and thorium nitrate proved that they both must be placed in the Y Inhalation Category of the ICRP lung model. It was therefore consistent that urinalysis and fecal analysis segments of the study were shown as not early indicators of significant lung uptakes of

Th^{232}. One of the most sensitive methods of detecting uptakes appears to be thoron in breath monitoring. Unfortunately in this study too few subjects had burdens measureable by other means for cross correlation. Information regarding the particle size is necessary to improve the accuracy of assigning lung burdens on the basis of thoron in breath. It is recommended that a sample of the thorium dioxide be analysed for particle size distribution, however, it is not recommended that thorium nitrate be analysed this way because of the physical change it undergoes in the lung.

The results of the monitoring at both facilities were consistent with bioassy measurements. The work environments that were identified as high potential for thorium inhalation were the work areas of the subjects with measureable lung burdens. There was a distinct difference between the potential risks at the two facilities. At the facility that used thorium dioxide there was no risk from thoron daughters in air because of the small inventory and tight binding of the thoron gas in the thorium dioxide particle. The material was very fine and does present a high potential for inhalation. The addition of a glove box at the coating stage and a change to wet instead of dry transfers to the ball mill will reduce the uptakes significantly. Education for and improvements in operator hygiene specifically regarding smoking and hand contamination should further reduce uptakes.

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FIGURE 4.1-1

Calibration Curve of Sodium Iodide Detector Using Ra226

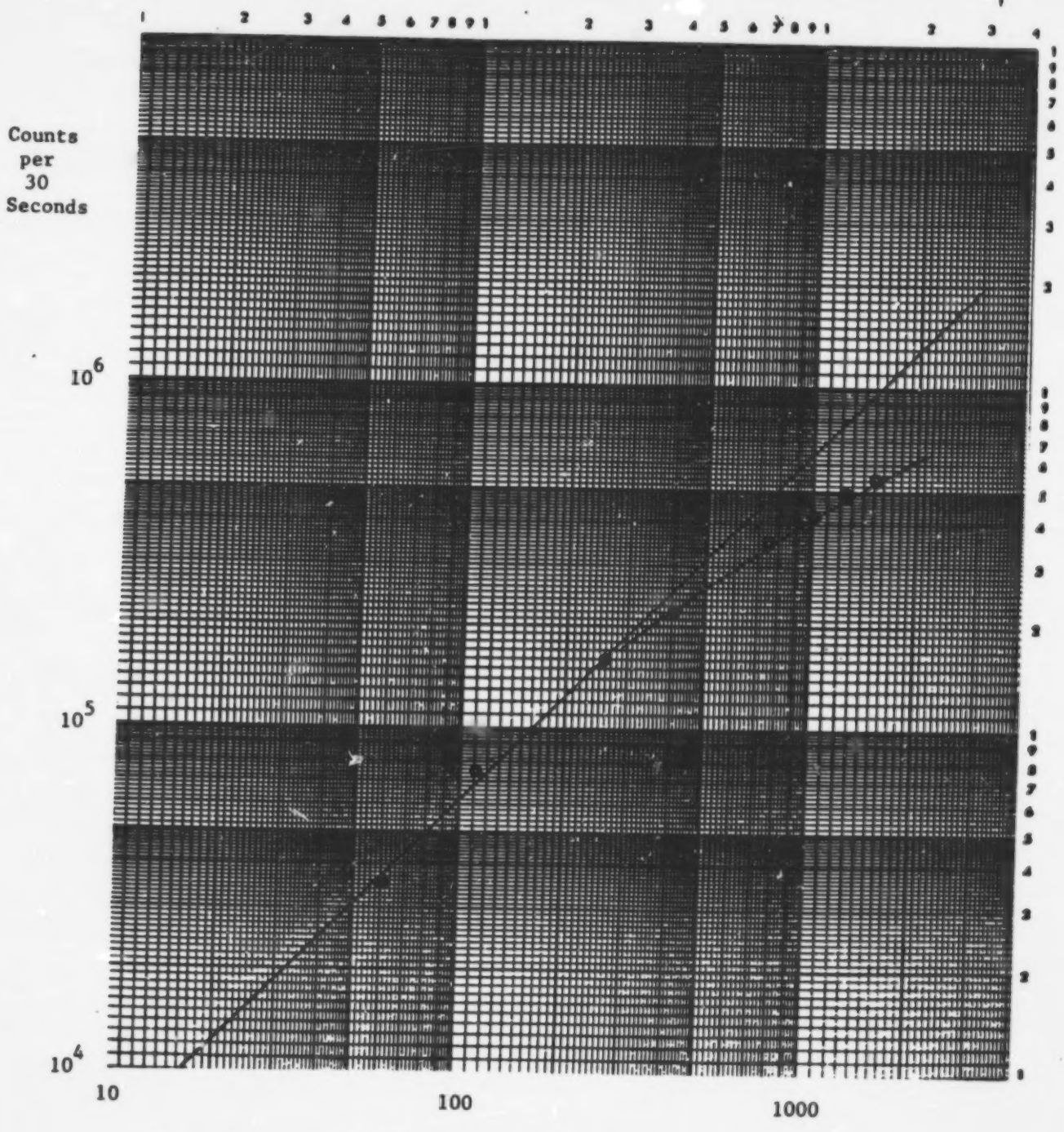


Figure 4.1-2

Calibration Curve for Sodium Iodide Detector (Ra226)

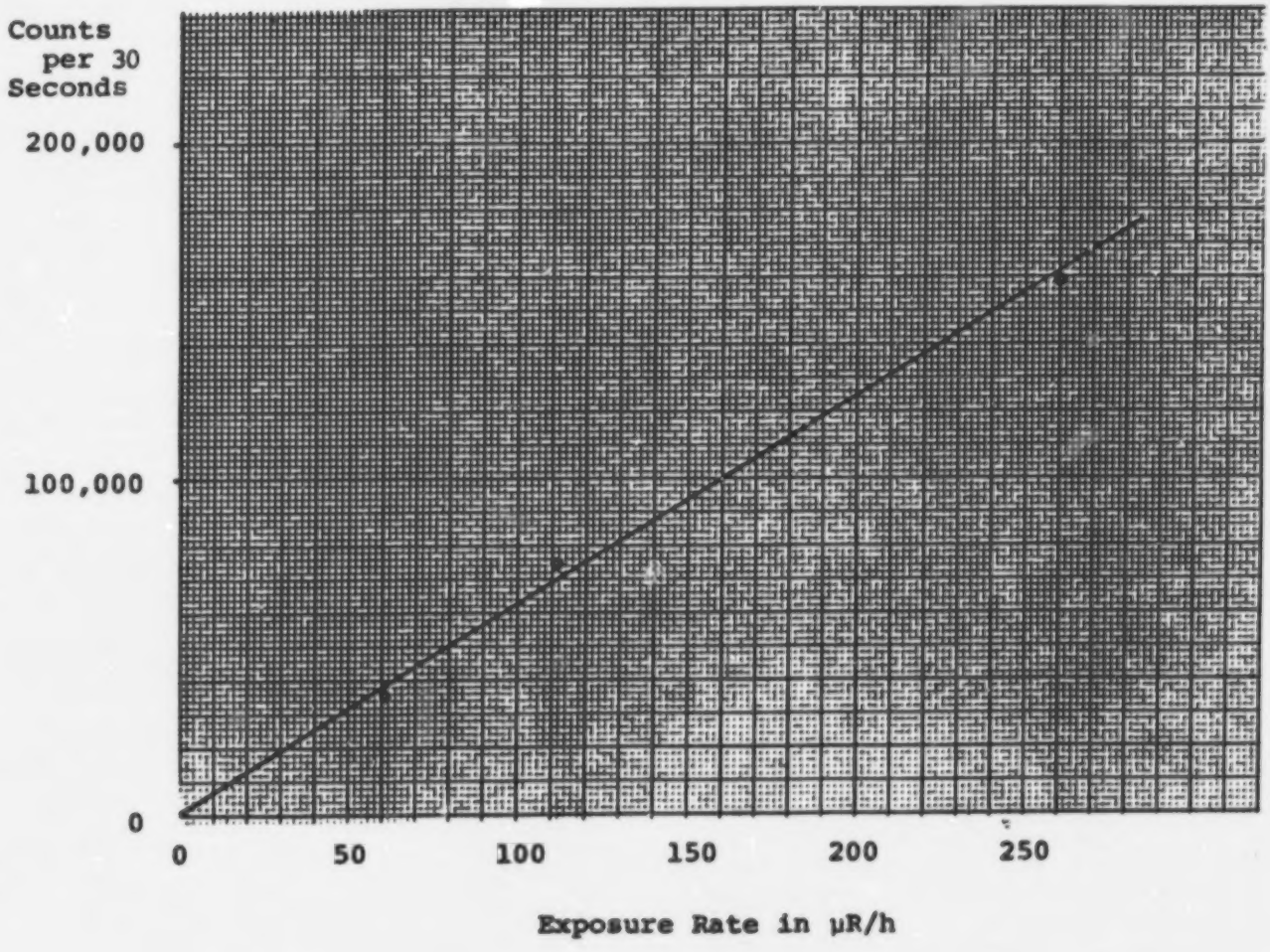
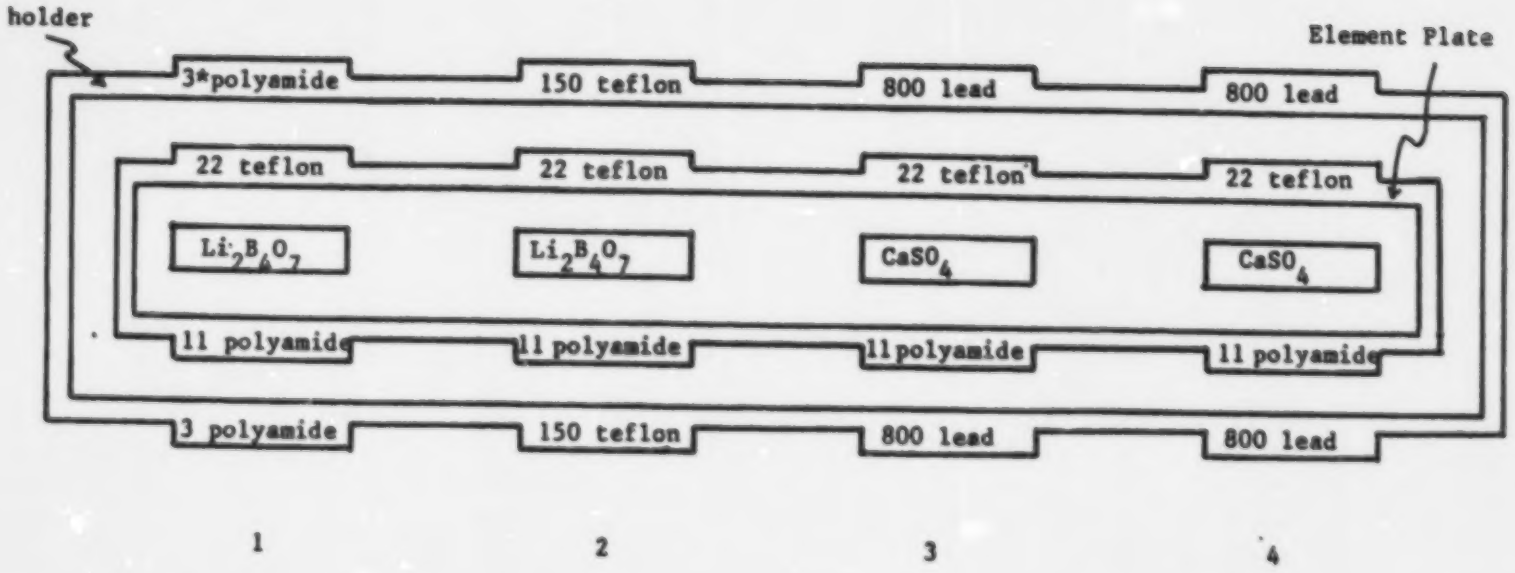


FIGURE 4.1-3

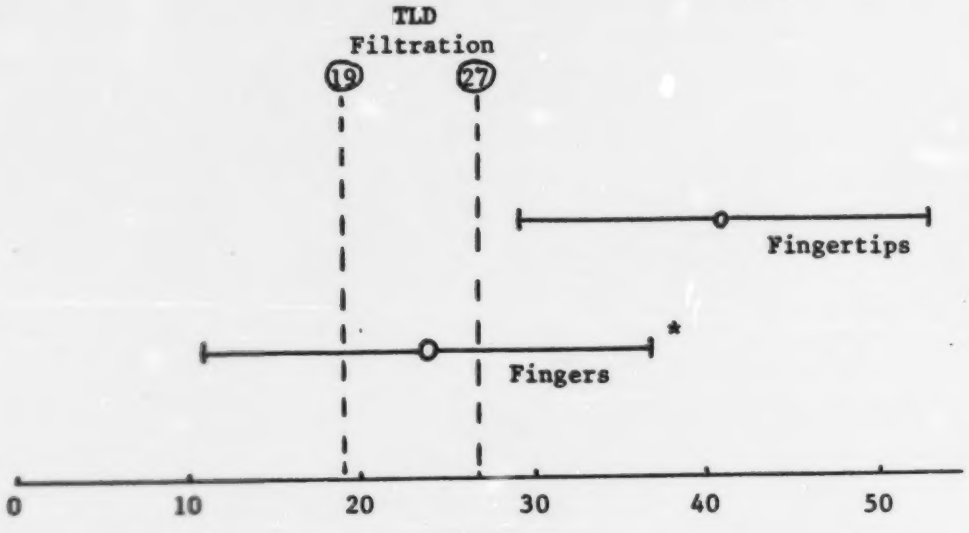
Schematic of Panasonic UD801 Thermoluminescent Dosimeter



* Units are mg/cm²

FIGURE 4.1-4

EPIDERMAL THICKNESS OF FINGERS AND TLD FILTRATION



Epidermal Thickness/TLD Filtration mg/cm²

* ± one standard deviation, taken from [ICRP23]

FIGURE 4.1-5

ANEMOMETER CALIBRATION - HIGH RANGE

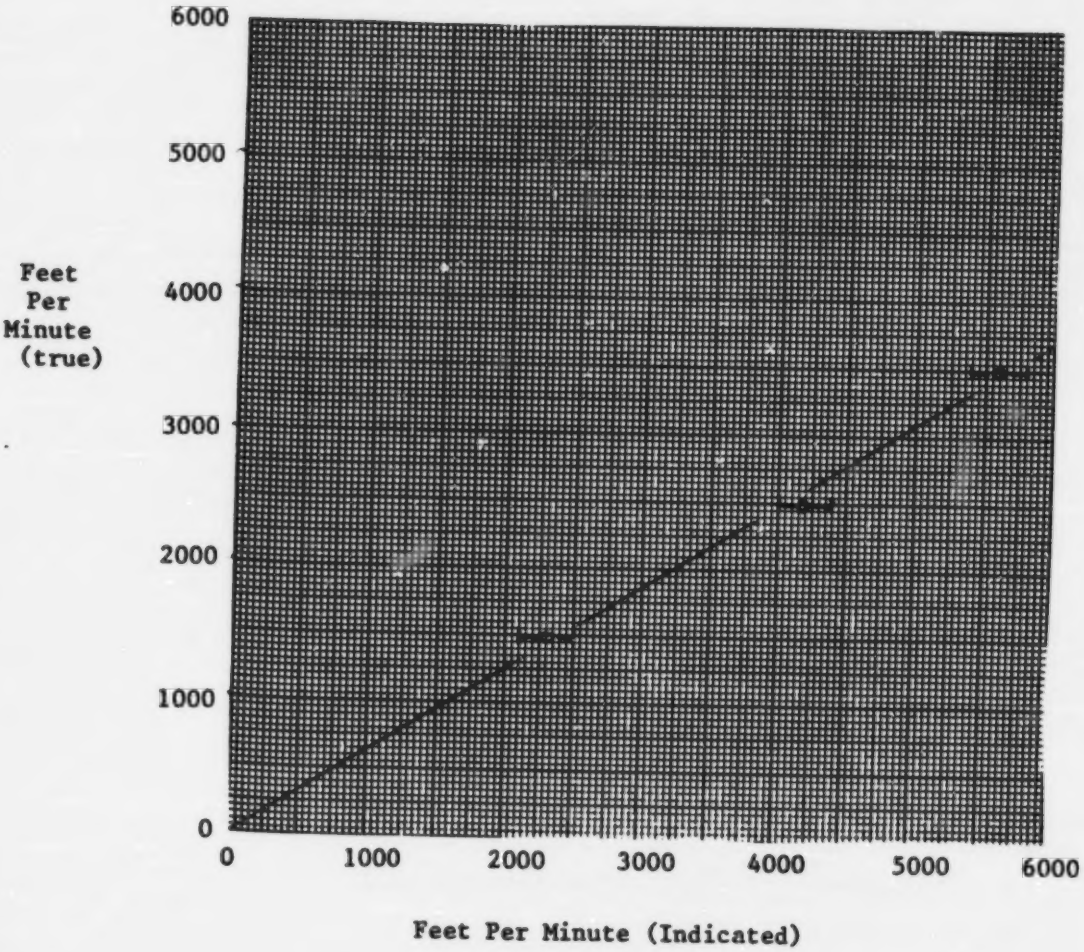


FIGURE 4.1-6

ANEMOMETER CALIBRATION - LOW RANGE

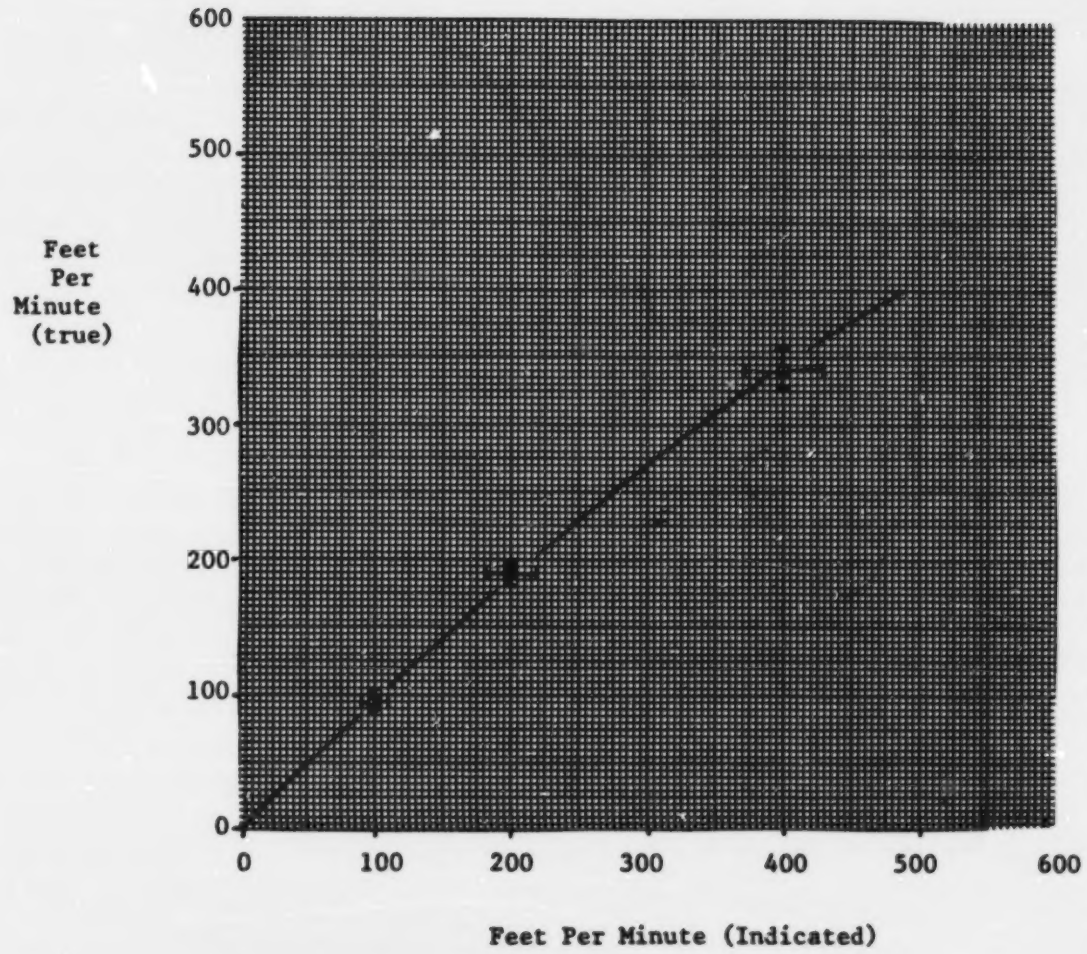


FIGURE 4.1-7

Th 228/Th 232 ACTIVITY RATIO AS A FUNCTION OF TIME SINCE SEPARATION

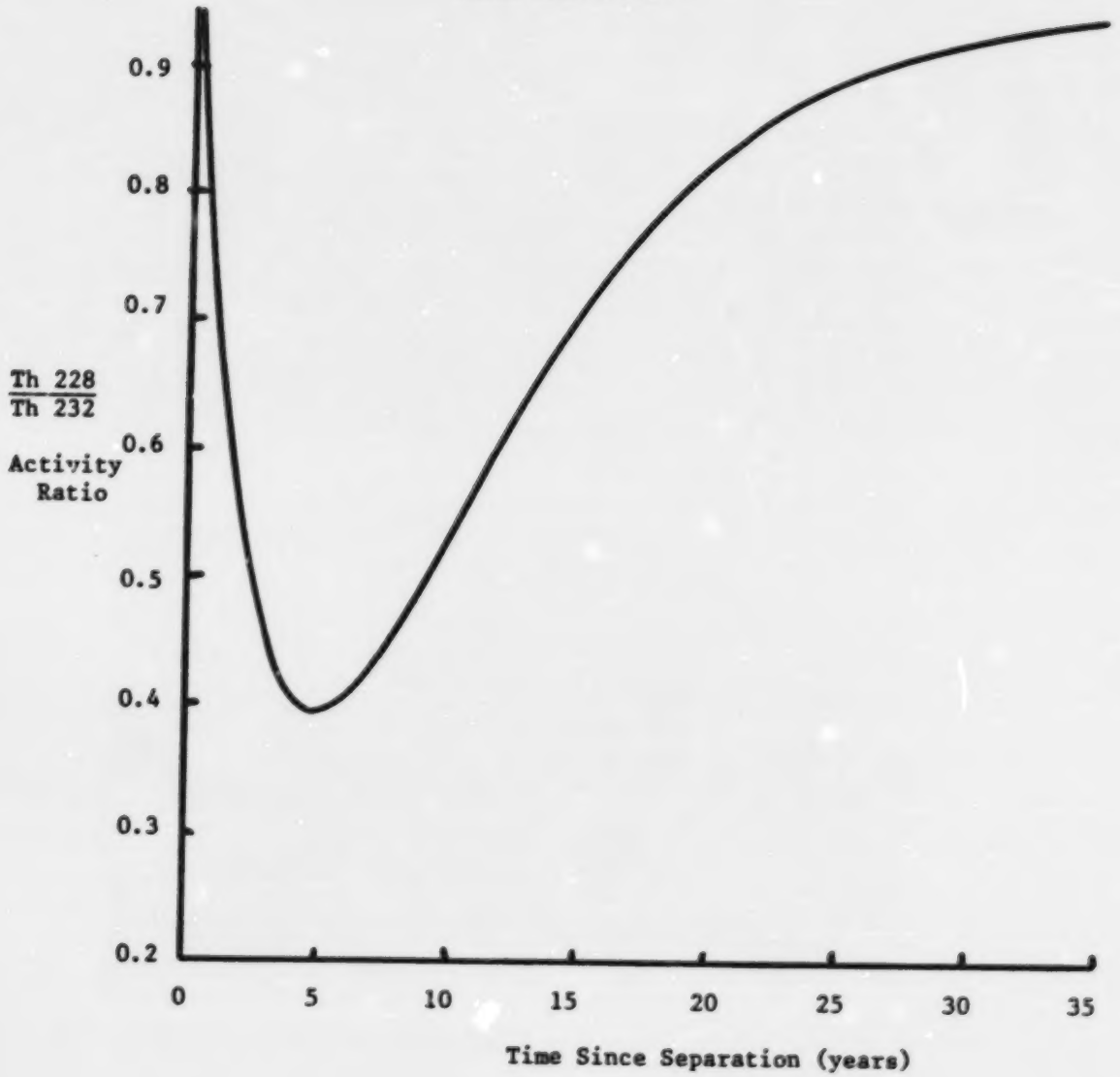
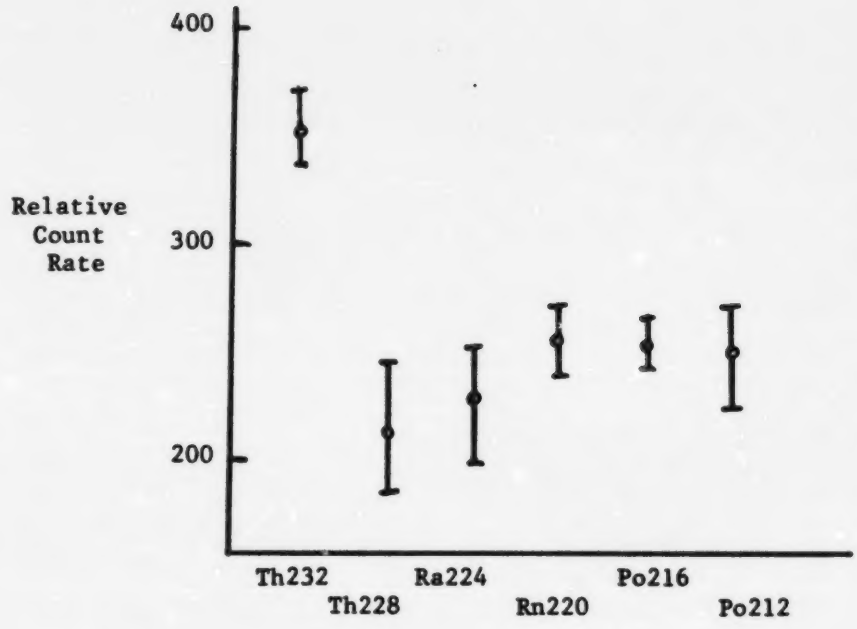


FIGURE 4.1-8

RELATIVE ACTIVITIES OF DAUGHTERS IN THORIUM DIOXIDE

STOCK MATERIAL FROM FACILITY B



± 2 standard deviations (counting statistics)

* divided by branching ratio of Po212 so actually represents Bi212 activity

FIGURE 4.2-1

GENERAL LAYOUT OF FACILITY A

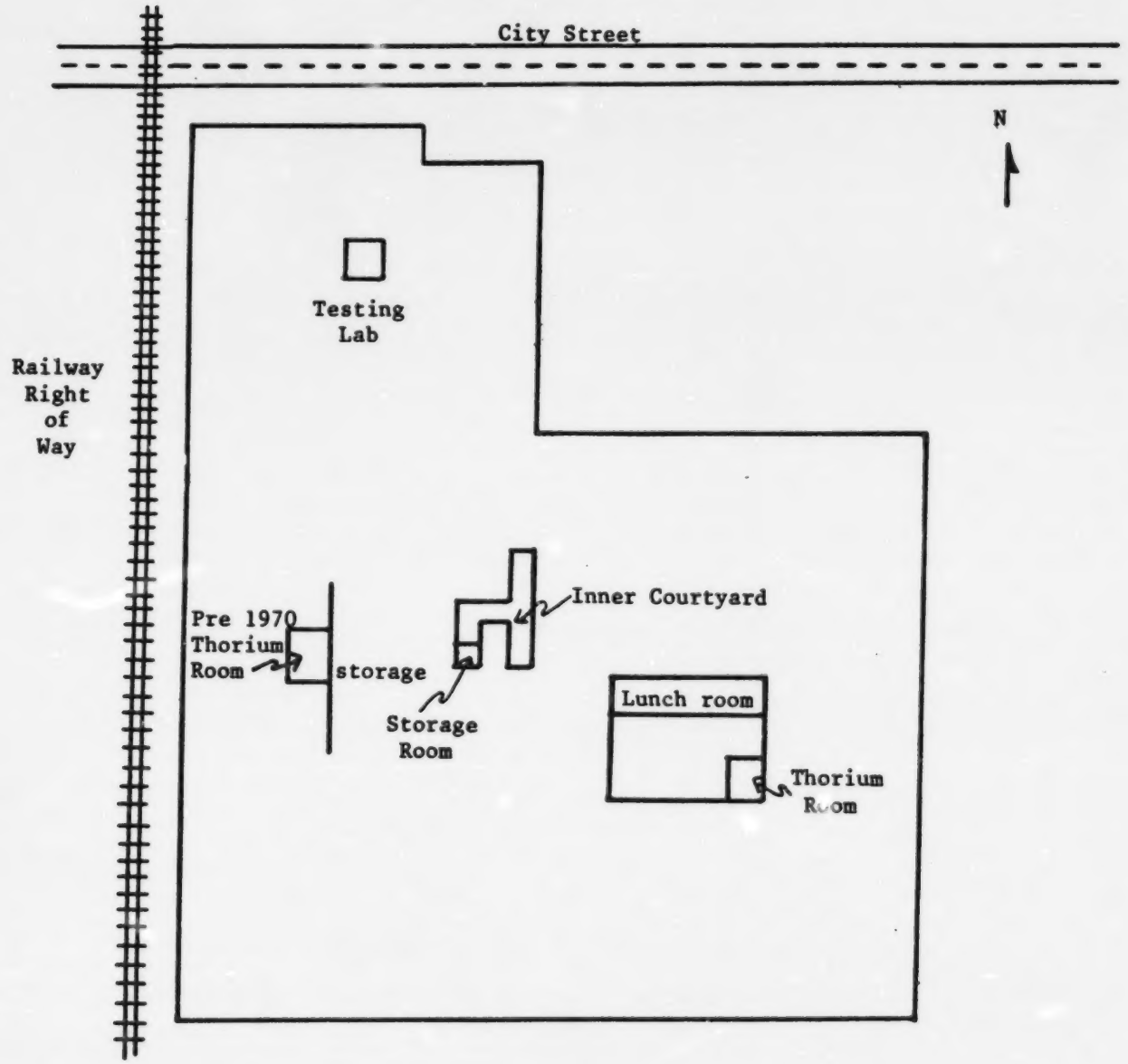


FIGURE 4.2-2

LAYOUT IN VICINITY OF THORIUM ROOM - FACILITY A

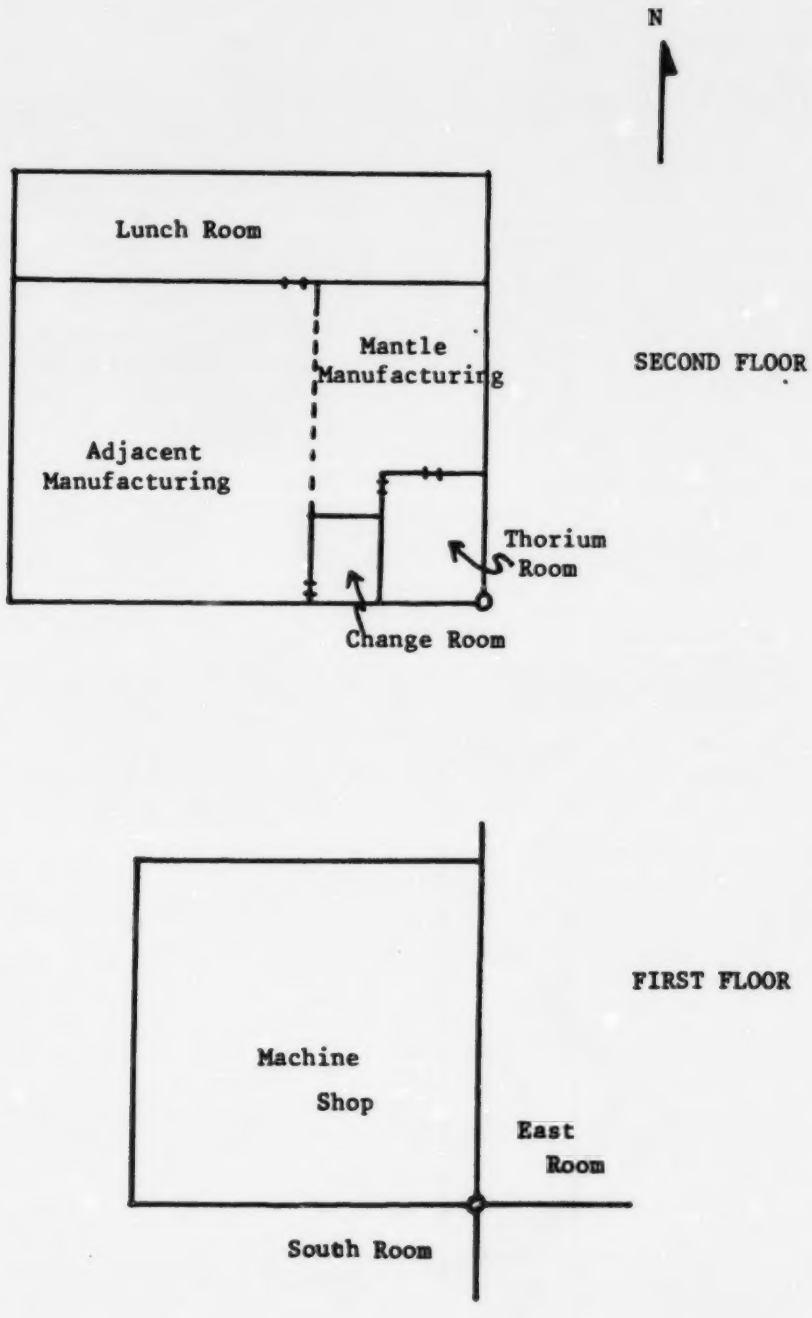


FIGURE 4.2-3

INNER COURTYARD COMPLETELY SURROUNDED BY BUILDINGS

FACILITY A

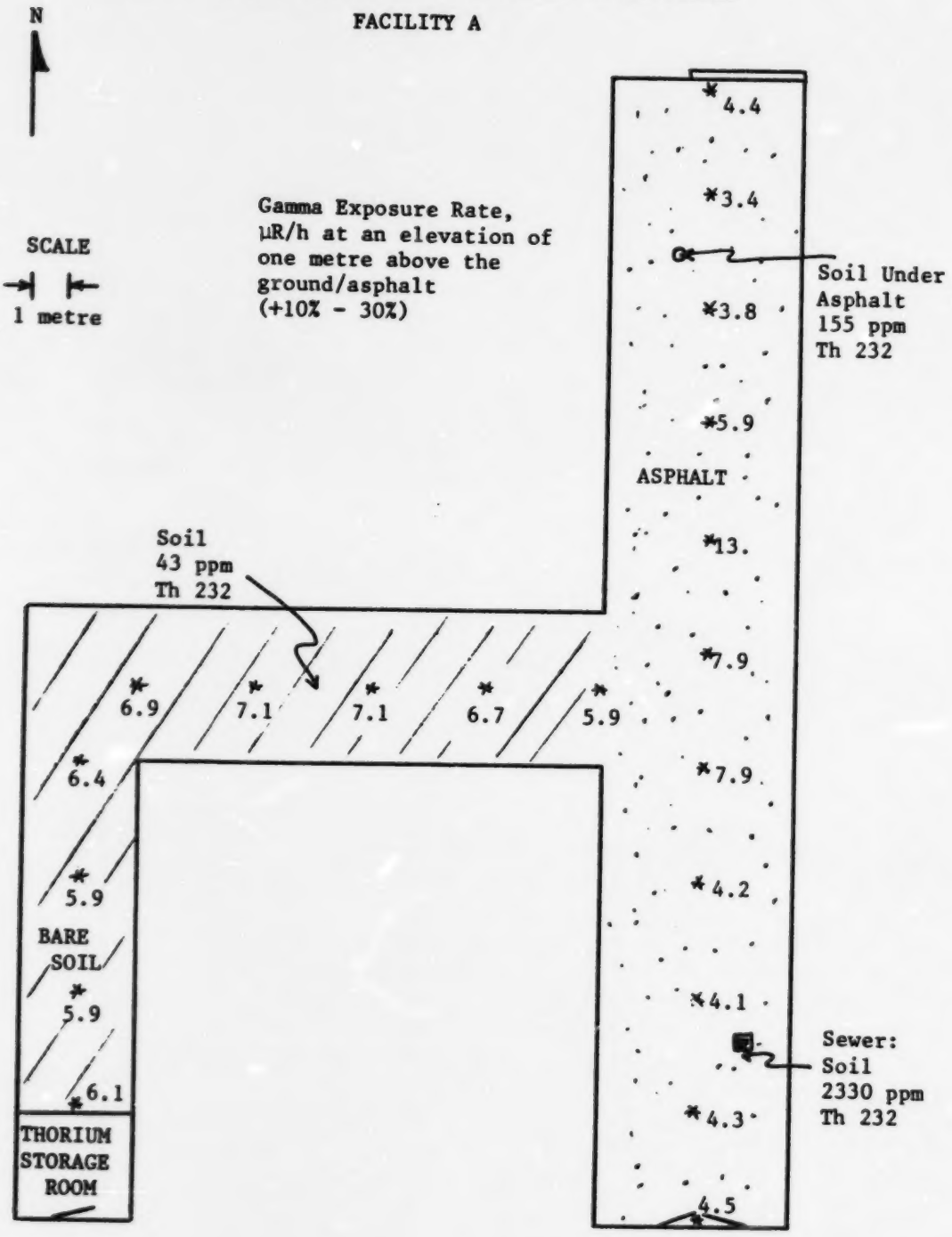
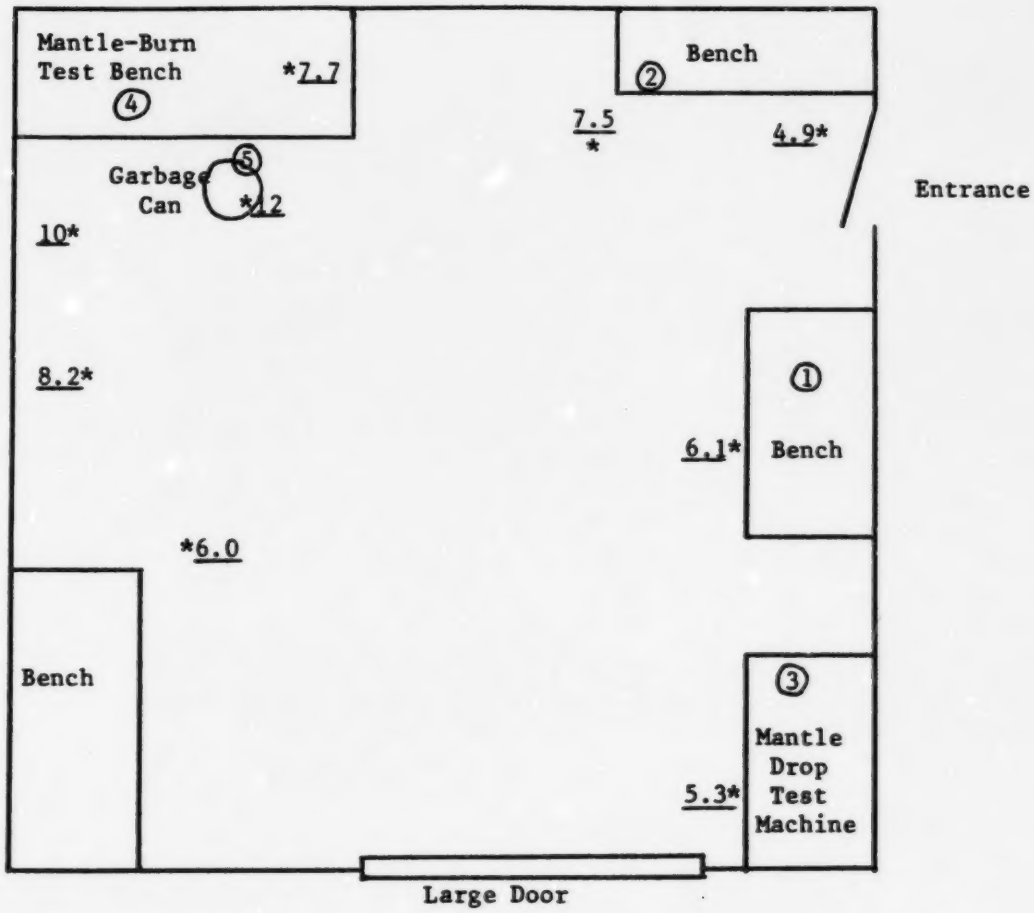


FIGURE 4.2-4

TEST LABORATORY FACILITY A: OCCUPATIONAL ENVIRONMENT MONITORING



Index - underlined numbers are exposure rates in μR/h at *

- circled numbers indicated locations where wipes were taken - removable alpha, dpm/100 cm²

① <10 dpm/100 cm²

② 10 ± 10 dpm/100 cm²

③ 3370 ± 180 dpm/100 cm²

④ 1040 ± 100 dpm/100 cm²

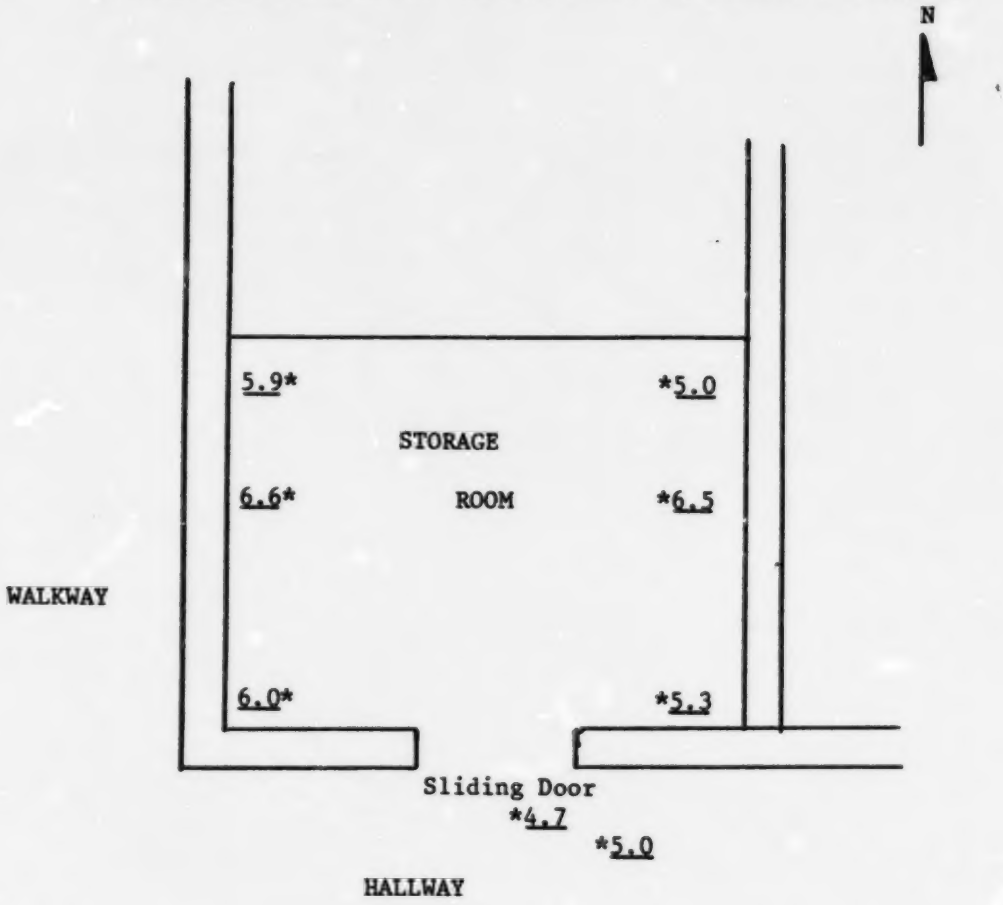
⑤ 490 ± 70 dpm/100 cm²

Thoron daughters is air: 1.7 ± 0.51 Bq/m³

0.8 ± 0.22 Bq/m³

FIGURE 4.2-5

STORAGE ROOM FACILITY A: OCCUPATIONAL ENVIRONMENT MONITORING



- underlined numbers are exposure rates in μR/h at *

- Thoron daughters in air inside the Storage Room
  0.4 ± 0.2 Bq/m³

FIGURE 7-1

FRACTION OF ANNUAL DOSE LIMIT (5 mSv/y) FOR NON ATOMIC RADIATION WORKERS

(assuming 2000 h/year occupancy)

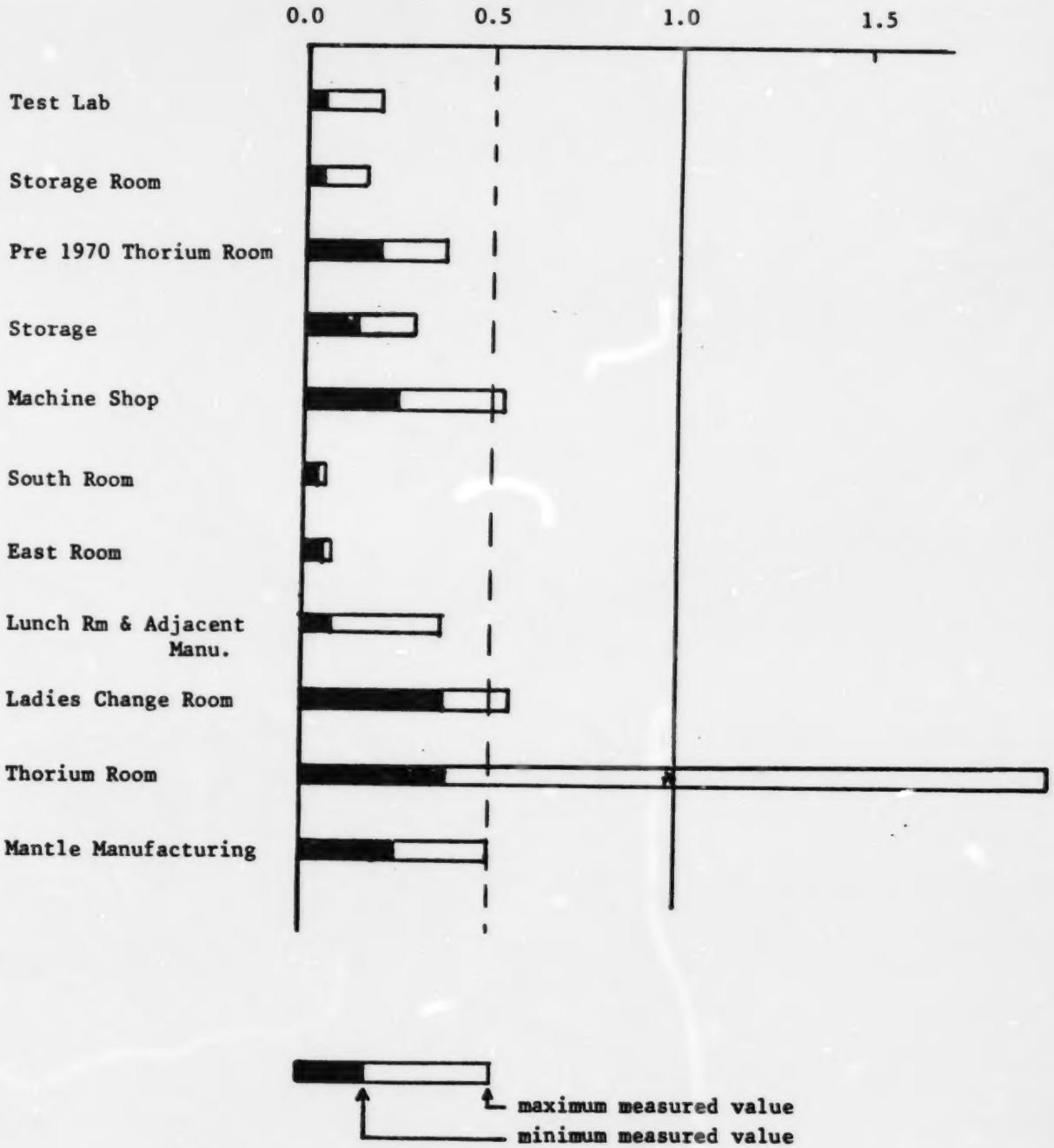


FIGURE 7-2

TH232 PATHWAYS FROM SOIL TO MAN

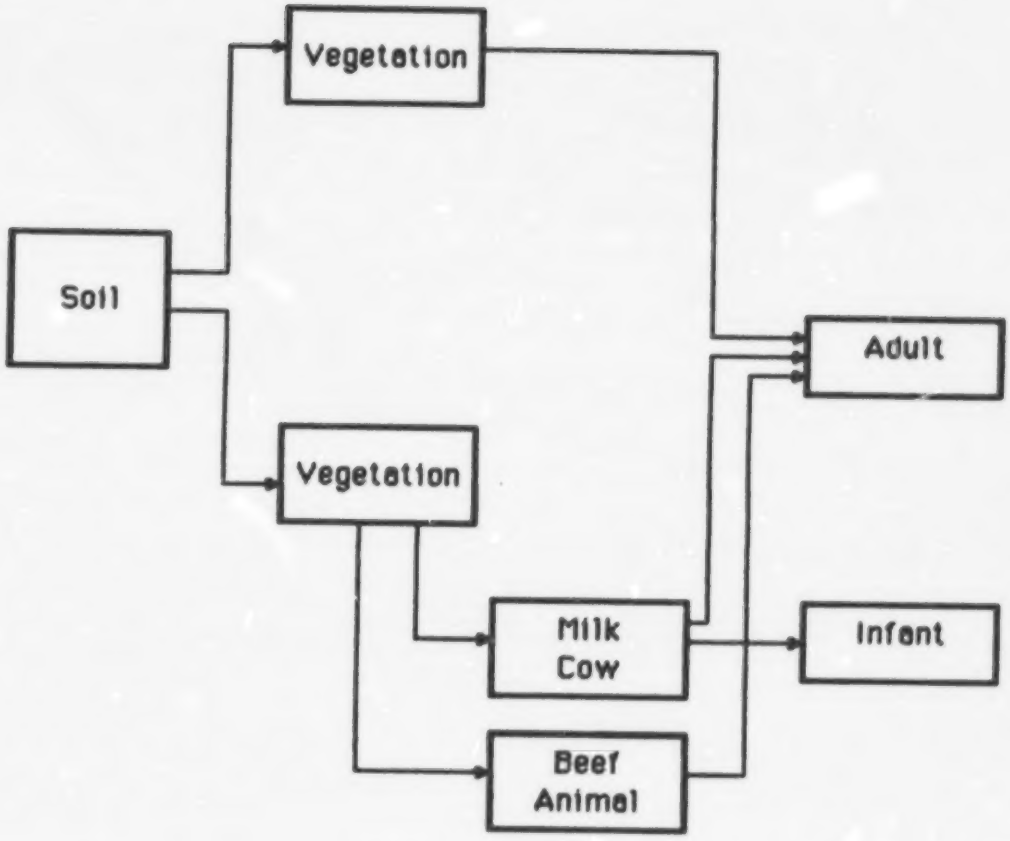


TABLE 2-1

JOB DESCRIPTIONS AND WORK HISTORIES OF FACILITY A SUBJECTS

Subject Number	Job Description* and Work History
A1	1950 to 1983
A2	1973 to 1983
A3	1973 to 1983
A4	1967 to 1983
A5	1969 to 1983
A6	1955 to 1983
A7	1966 to 1983
A8	maintenance on equipment 1979 to 1983
A9	1964 to 1983

* All subjects (except A8) rotated through all the tasks except for mixing of the solutions and dipping of the mantle materials.

TABLE 2-2

JOB DESCRIPTIONS AND WORK HISTORIES OF FACILITY B SUBJECTS

Subject #	Job Description and Work History
B1	August 2, 1972 to present, 8 years on quartz lathe 1 year odd jobs, on thorium press to present
B2	September 1983 to present, electrolytic cleaning and cutting of thorium metal and inspects electrodes for thorium deposition
B3	May 15, 1972 to present, coating room for 11 years to December 1983, since then occasionally fills in at coating room
B4	October 30, 1972 to present, on thorium presses for 9 years, for 2 years occasional work on thorium presses
B5	September 1968 to present, welding coils to hairpins
B6	November 6, 1972 to November 11, 1974 and May 18, 1981 to present, odd jobs and cleaning out radwaste from coating room
B7	April 11, 1966 to present, only person to work with thorium in mixing room
B8	December 1983 to present, works in coating room
B9	February 5, 1979 to present, mechanic maintains equipment in all thorium areas except the mixing room
B10	Started in coating room August 1975, maternity leave January - October 1980 and May 1983 to May 1984
B11	April 19, 1972 to present, all tasks except mixing room on a replacement basis

TABLE 2-3

THORIUM DIOXIDE USAGE AT FACILITY B SINCE 1966

Year	Batches	
1967	1	
1968	11	
1969	15	
1970	18	
1971	22	each batch contained 989 g of ThO_2
1972	36	
1973	53	
1974	14	
1975	7	each batch contained 1978 g of ThO_2
1976	7	
1977	7	
1978	9	
1979	8	
1980	5	
1981	5	
1982	3	
1983	3.5	

TABLE 3-1

THORIUM 232 DECAY CHAIN

Nuclide	Historical name	Half-life	Major radiation energies (MeV) and intensities†		
			α	β	γ
$^{232}_{90}Th$	Thorium	1.41×10^{10}y	3.95 (24%) 4.01 (76%)	---	---
$^{228}_{88}Ra$	Mesothorium I	6.7y	---	0.055 (100%)	---
$^{228}_{89}Ac$	Mesothorium II	6.13h	---	1.18 (35%) 1.75 (12%) 2.09 (12%)	0.34c‡ (15%) 0.908 (25%) 0.96c (20%)
$^{228}_{90}Th$	Radiothorium	1.910y	5.34 (28%) 5.43 (71%)	---	0.084 (1.6%) 0.214 (0.3%)
$^{224}_{88}Ra$	Thorium X	3.64d	5.45 (6%) 5.68 (94%)	---	0.241 (3.7%)
$^{220}_{86}Rn$	Emanation Thoron (Tn)	55s	6.29 (100%)	---	0.55 (0.07%)
$^{216}_{84}Po$	Thorium A	0.15s	6.78 (100%)	---	---
$^{212}_{82}Pb$	Thorium B	10.64h	---	0.346 (81%) 0.586 (14%)	0.239 (47%) 0.300 (3.2%)
$^{212}_{83}Bi$	Thorium C	60.6m	6.05 (25%) 6.09 (10%)	1.55 (5%) 2.26 (55%)	0.040 (2%) 0.727 (7%) 1.620 (1.8%)
64.0% 36.0%					
$^{212}_{84}Po$	Thorium C'	304ns	8.78 (100%)	---	---
$^{208}_{81}Tl$	Thorium C''	3.10m	---	1.28 (25%) 1.52 (21%) 1.80 (50%)	0.511 (23%) 0.583 (86%) 0.860 (12%) 2.614 (100%)
$^{208}_{82}Pb$	Thorium D	Stable	---	---	---

Data taken from: Lederer, C. M., Hollander, J. M., and Perlman, I., Table of Isotopes (6th ed.; New York: John Wiley & Sons, Inc., 1967) and Hagen, O. H., Zigman, P. E., and Mechin, J. L., Beta Spectra (UCRL-TR-802 [Washington, D.C.: U.S. Atomic Energy Commission, 1964]).

TABLE 4.2-1

THORIUM ROOM EXHAUST STACKS - FACILITY A

Exhaust Vent	Dimensions in x in	Air Speed FPM	Maximum Exhaust Rate m^3/s	Duty Cycle Shift h/8h	Duty Cycle Week * h/168h	Average Exhaust Rate $x10^3 m^3/8h$	Average Exhaust Rate $x10^3 m^3/168h$*
Dryer Exhaust	16 x 12	1600	1.01	8	40	29	145
Lacquer Exhaust	16 x 12	2200	1.38	7	35	35	175
Ammonia Exhaust	7.75 dia	800	0.124	4 ± 2	20 ± 10	1.79	9.0
Old Thorium Room	12 x 9	570	0.20	0 - 8	0 - 40	0 - 5.8	0 - 29
New Thorium Room	13.5 x 9	1350	0.54	0 - 8	0 - 40	0 - 15.6	0 - 78
					TOTALS	66 - 87	330 - 436

* Assuming One Shift Per Day Each of 5 Days Per Week

TABLE 4.2-2

CONTACT BETA-GAMMA EXPOSURE RATE ON LANTERN MANTLES

TLD No.	Total Filtration mg/cm²		Exposure Rate mR/h	Average mR/h
		MANTLE 'X'		
98	19		5.2	4.8
97	19		4.5	
96	27		4.1	4.2
95	27		4.3	
93	19	+	3.5	3.9
92	19	Cotton	4.2	
91	19	Glove	4.1	
90	27	+	3.8	3.5
89	27	Cotton	3.1	
88	27	Glove	3.4	
		MANTLE'Y'		
86	19		5.0	4.6
85	19		4.1	
83	27		3.6	3.8
82	27		3.9	
81	19	+	4.2	4.0
80	19	Cotton Glove	3.7	
78	27	+	3.6	3.5
77	27	Cotton	3.3	
72	27	Glove	3.6	

TABLE 4.2-3

CONTACT BETA-GAMMA EXPOSURE RATE ON THORIUM METAL & ELECTRODES

TLD No.	Total Filtration mg/cm²		Exposure Rate mR/h	Average mR/h
		THORIUM METAL		
70	19		31	30
71	19		30	
69	19	+ Cotton Glove	16.6	17.3
68	19		18.1	
67	27		18.5	18.5
65	27		18.5	
64	27	+ Cotton Glove	17.4	18.6
63	27		19.8	
		ELECTRODES		
		(one small & one large electrode on each chip)		
62	19		0.57	0.63
61	19		0.70	
60	19	+ Cotton Glove	0.36	0.45
59	19		0.55	
57	27		0.10	0.34
56	27		0.59	
55	27	+ Cotton Glove	0.25	0.23
54	27		0.20	

TABLE 5.2-1

SUMMARY OF BIOASSY RESULTS OF FACILITY A SUBJECTS

Subject	Whole Body Bq Th232 (Tl208)		Lung Bq Th232 (Pb212)		Thoron in Breath mBq Rn220/min		Urinanlysis - output mBq/24 h					
							Gross alpha (Th)		Ra224		Ra228	
	March	June	March	June	March	June	March	August	March	August	March	August
A1	ND	--	ND	--	64 ± 32	--	<0.19	0.08	-(1)	1.12	-(1)	4.3
A2	ND	--	ND	--	62 ± 32	--	6.9	0.147	--	0.66	--	5.7
A3	ND	--	ND	--	52 ± 28	--	0.19	0.21	--	0.67	--	1.2
A4	ND	--	ND	--	62 ± 32	--	0.48	0.174	--	1.07	--	11.0
A5	ND	--	ND	--	28 ± 20	--	0.80	0.35	--	2.7	--	49.0
A6	14 ± 14	68 ± 133	31 ± 22	36 ± 26	271 ± 86	300 ± 96	9.0	1.3	--	4.2	--	<1.0
A7	ND	ND	ND	ND	109 ± 46	135 ± 52	2.6	0.44	--	0.39	--	10.8
A8	ND	--	ND	--	58 ± 30	--	0.54	0.84	--	1.17	--	39.0
A9	ND	ND	ND	ND	82 ± 38	48 ± 28	2.7	0.10	--	1.20	--	<1.0
						(4)		(2)		(3)		(3)

NOTES: (1) contaminated resin
(2) normal urinary excretion rate of thorium is 0.4 mBq/day [ICRP23]
(3) normal urinary excretion rate of Ra226 is 3.0 mBq/day [ICRP23]
(4) 80 mBq/min is considered upper limit of normal exhalation rate

TABLE 5.2-2

THORON IN BREATH RESULTS - FACILITY A

Subject	Net Counts 170 - 1010 min		Thoron in Breath mBq Rn220/min	
	March	June	March	June
A1	32 ± 16	--	64 ± 32	--
A2	31 ± 16	--	62 ± 32	--
A3	26 ± 14	--	52 ± 28	--
A4	31 ± 16	--	62 ± 32	--
A5	14 ± 10	--	28 ± 20	--
A6	136 ± 43	153 ± 48	271 ± 86	300 ± 96
A7	55 ± 23	68 ± 26	109 ± 46	135 ± 52
A8	29 ± 15	--	58 ± 30	--
A9	41 ± 19	24 ± 14	82 ± 38	48 ± 28

TABLE 5.2-3

March Urinalysis Results - Facility A

Subject	Urine Collection Period (hours)	Gross Alpha Activity in Urine[c] mBq/sample	mBq/24 hours
A1	25	< 0.19	< 0.19
A2	18	5.2	6.9
A3	32	0.25	0.19
A4	30	0.60	0.48
A5	24	0.80	0.80
A6	15	5.6	9.0
A7	18.5	2.0	2.6
A8	25.5	0.57	0.54
A9	21	2.4	2.7

Note: No radium results due to contamination

[c] Alpha corrected to pure Th232/Th228, α spectrum shows very small Th 230 contribution.

TABLE 5.2-4

AUGUST URINALYSIS RESULTS - FACILITY A

Subject	Urine Collection Period (hours)	Radioactivity in Urine mBq/sample α	Ra224	Ra228	mBq/24 hours α	Ra224	Ra228
A1	24	0.08	1.12	4.3	0.08	1.12	4.3
A2	24.5	0.15	0.67	5.8	0.147	0.66	5.7
A3	25	0.22	0.70	1.3	0.21	0.67	1.2
A4	23.5	0.17	1.05	10.9	0.174	1.07	11.0
A5	24	0.35	2.7	48.9	0.35	2.7	49.0
A6	24	1.3	4.2	0.0	1.3	4.2	<1.0
A7	23	0.42	0.37	10.4	0.44	0.39	10.8
A8	23	0.08	1.12	37.4	0.84	1.17	39.0
A9	24	0.10	1.20	0.0	0.10	1.20	<1.0

TABLE 5.2-5

MARCH FECES RESULTS - FACILITY A

Subject	Feces Collection period (hours)	Radioactivity in Feces mBq/sample			mBq/24 hours		
		α	Ra (e)	Ra228	α	Ra (e)	Ra228
A1	24	29.2	4.5	9.3	29.2	4.5	9.3
A2	24	15.5	13.2	0.0	15.5	13.2	<7.0
A3	24	27.0	13.3	25.0	27.0	13.3	25.0
A4	24	13.7	20.7	7.1	13.7	20.7	7.1
A5	24	27.3	69.5	0.0	27.3	69.5	<7.0
A6	24	10.8	41.5	160.0	10.8	41.2	160.0
A7	16	28.3	40.5	0.0	42.5	61.0	<7.0
A8	15	33.5	33.8	0.0	54.0	54.0	<7.0
A9	24	46.3	15.7	85.0	46.3	15.7	85.0

(e) mainly radium 226

TABLE 5.2-6

Th 232 IN HAIR AND NAILS - SUBJECTS FROM FACILITY A

Subject	Th 232 Content in Parts per Million	
	Toe & Finger Nails	Hair
A1	11.9 (±5%)	0.3 (±36%)
A3	3.9 (±13%)	< 0.3
A5	< 0.8	< 0.24
A6	4.8 (±9.6%)	13.8 (±4.1%)
A7	9.5 (±8.3%)	0.3 (±37%)
A9	2.0 (±11%)	< 0.19

TABLE 5.3-1

SUMMARY OF BIOASSY RESULTS OF FACILITY B SUBJECTS

Subject	Whole Body Bq Th232 (Tl208)		Lung Bq Th232 (Pb212)		Thoron in Breath mBq Rn220/min		Urinalysis - output mBq/24 hours Gross alpha (Th)		Ra224		Ra228	
	March	August	March	August	March	August	March	August	March	August	March	August
B1	ND	--	ND	--	68 ± 34	--	0.0	0.0	0.0	1.22	<1.0	<1.0
B2	ND	ND	ND	ND	167 ± 62	30 ± 12	0.0	0.13	0.0	2.3	0.89	<1.0
B3	ND	ND	22 ± 22	20 ± 24	240 ± 80	290 ± 92	0.044	0.0	0.0	2.0	0.39	<1.0
B4	ND	--	ND	--	36 ± 22	--	0.077	0.13	1.23	0.40	<1.0	3.3
B5	ND	--	ND	--	76 ± 36	--	0.0	0.35	1.6	1.7	0.89	<1.0
B6	ND	ND	ND	ND	82 ± 38	113 ± 32	0.26	0.20	0.0	1.8	<1.0	14.3
B7	ND	ND	ND	ND	125 ± 50	117 ± 34	0.0	0.10	4.6	4.2	28.0	<1.0
B8	ND	ND	ND	ND	30 ± 20	74 ± 22	0.0	0.13	0.10	1.5	0.61	1.2
B9	ND	--	ND	--	40 ± 24	--	0.30	0.18	0.0	0.36	3.0	18.0
B10	--	ND	--	29 ± 24	--	610 ± 125	--	0.067	--	25.0	--	<1.0
B11	ND	--	ND	--	34 ± 22	--	0.0	0.017	1.78	0.73	73.0	16.3
						(3)	(1)		(2)		(2)	

NOTES: (1) normal urinary excretion rate of thorium is 0.4 mBq/day [ICRP23]
(2) normal urinary excretion rate of Ra226 is 3.0 mBq/day [ICRP23]
(3) 80 mBq/min is considered upper limit of normal exhalation rate

TABLE 5.3-2

THORON IN BREATH - FACILITY B

Subject	Net Counts 170 - 1010 min		Thoron in Breath mBq Rn220/min	
	March	August	March	August
B1	34 ± 17	--	68 ± 34	--
B2	84 ± 31	15 ± 6	167 ± 62	30 ± 12
B3	122 ± 40	144 ± 46	240 ± 80	290 ± 92
B4	18 ± 11	--	36 ± 22	--
B5	38 ± 18	--	76 ± 36	--
B6	41 ± 19	57 ± 16	82 ± 38	113 ± 32
B7	63 ± 25	59 ± 17	125 ± 50	117 ± 34
B8	15 ± 10	37 ± 11	30 ± 20	74 ± 22
B9	20 ± 12	--	40 ± 24	--
B10	--	305 ± 63	--	610 ± 125
B11	17 ± 11	--	34 ± 22	--

TABLE 5.3-3

MARCH URINALYSIS RESULTS - FACILITY B

Subject	Urine Collection Period (hours)	Radioactivity in Urine mBq/sample α	Ra224	Ra228	mBq/24 hours α	Ra224	Ra228
B1	22	0.0	0.0	0.0	0.0	0.0	<1.0
B2	29	0.0	0.0	1.07	0.0	0.0	0.89
B3	27.5	0.05	0.0	0.45	0.044	0.0	0.39
B4	25	0.08	1.28	0.0	0.077	1.23	<1.0
B5	19.5	0.0	1.3	0.72	0.0	1.6	0.89
B6	25	0.27	0.0	0.0	0.26	0.0	<1.0
B7	17.3	0.0	3.3	20.0	0.0	4.6	28.0
B8	24.5	0.0	0.1	0.62	0.0	0.1	0.61
B9	12	0.15	0.0	1.5	0.3	0.0	3.0
B11	23	0.0	1.7	70.3	0.0	1.78	73.0

TABLE 5.3-4

AUGUST URINALYSIS RESULTS - FACILITY B

Subject	Urine Collection Period (hours)	Radioactivity in Urine mBq/sample α	Ra 224	Ra228	mBq/24 hours α	Ra224	Ra228
B1	25.5	0.0	1.3	0.0	0.0	1.22	<1.0
B2	24	0.13	2.3	0.0	0.13	2.3	<1.0
B3	24	0.0	2.0	0.0	0.0	2.0	<1.0
B4	23.25	0.13	0.40	3.4	0.13	0.40	3.3
B5	24	0.35	1.7	0.0	0.35	1.7	<1.0
B6	24	0.20	1.8	14.3	0.20	1.8	14.3
B7	23	0.10	4.0	0.0	0.10	4.2	<1.0
B8	27	0.15	1.7	1.4	0.133	1.5	1.2
B9	20	0.15	0.30	15.2	0.18	0.36	18.0
B10	24	0.067	25.0	0.0	0.067	25.0	<1.0
B11	23.5	0.017	0.73	0.62	0.017	0.73	0.63

TABLE 5.3-5

MARCH FECES RESULTS - FACILITY B

Subject	mBq Excreted Per 24 Hours		
	α	Ra224	Ra228
B1	9.8	< 1	30.
B2	27.0	35	< 8.
B3	13.0	1.2	92.5
B4	4.0	< 1	14.3
B5	35.	1.3	< 8.
B6	31.	42.	< 8.
B7	6.3	30.	7.7
B8	31.	42.	< 8.
B9	17.	23.	< 8.
B11	6.8	28.	< 8.

TABLE 5.3-6

TH232 IN HAIR AND NAILS - SUBJECTS FROM FACILITY B

Subject	Th 232 Content in Parts Per Million			
	Toe & Finger Nails		Hair	
B2	1.0	(±22%)	< 0.17	
B3	< 0.4		< 0.12	
B5	4.6	(±9.6%)	1.28	(±9%)
B6	2.8	(±7.2%)	< 0.13	
B7	0.4	(±45%)	< 0.5	
B10	11.7	(±8.7%)	0.56	(±13%)
B11	< 0.4		< 0.15	

TABLE 6-1

COMPOSITION OF SIMULATED LUNG FLUID

Salt	Simulated Lung Fluid SLF[c]
KCl	0.004
NaCl	0.145
$MgCl_2$	0.001
NH_4Cl	-
$NaHCO_3$	0.024
Glycine	-
L-Cysteine	-
Na_3Citrate	0.0003
Na Acetate	0.007
$CaCl_2$	0.0025
H_2SO_4	-
Na_2SO_4	0.0005
Na_2HPO_4	0.001
NaH_2PO_4	-
DTPA[d]	-
ABDC[e]	-

[a]Aqueous solution.

[b]E180.

[c]Kal79, De82.

[d]Diethylenetriaminepentaacetic acid, not present in blood serum.

[e]Alkylbenzyldimethylammonium chloride, an antibacterial agent not present in blood serum.

TABLE 6-2

SOLUBILITY OF THORIUM DIOXIDE FROM FACILITY B IN SIMULATED LUNG FLUID

Day	DISSOLVED CONCENTRATION IN Bq/L		
	Gross α	Uranium	Ra 224
1	0.123	0.167	63
3	0.032	0.167	35
10	0.027	0.167	18.3
30	0.045	0.150	12.2

TABLE 6-3

SOLUBILITY OF THORIUM IN THORIUM DIOXIDE FOR

FACILITY B IN SIMULATED LUNG FLUID

Day	Dissolution Rate	
	mBq/d	As Fraction of Undissolved per day
1	37.0	9.8×10^{-6}
3	9.6	2.6×10^{-6}
10	8.1	2.2×10^{-6}
30	3.5	3.6×10^{-6}

TABLE 6-4

SOLUBILITY OF RA224 IN THORIUM DIOXIDE FROM FACILITY B

IN SIMULATED LUNG FLUID

Day	Dissolution Rate	
	Bq/d	As Fraction of Undissolved per day
1	18.9	0.0084
3	10.5	0.0047
10	5.5	0.0024
30	3.7	0.0016

TABLE 7-1

FACTOR USED IN PATHWAY ANALYSIS CALCULATIONS

Radioisotope	food crop[1] concentration factors	adult dose factors
Th 232	5×10^{-4}	1.1×10^{-4}
Ra 228	4×10^{-2}	2.6×10^{-7}
Ac 228	1×10^{-3}	---
Th 228	5×10^{-4}	1.5×10^{-5}
Ra 224	4×10^{-2}	9.3×10^{-8}

(1) Bq/g fresh food per Bq/g dry soil from Ss57

(2) committed effective dose equivalent Sv/Bq from Jo83